

REPORT DOCUMENTATION PAGE			Form Approved OMB NO. 0704-0188		
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1. REPORT DATE (DD-MM-YYYY) 15-03-2022		2. REPORT TYPE Final Report		3. DATES COVERED (From - To) 1-Jul-2014 - 31-Dec-2021	
4. TITLE AND SUBTITLE Final Report: MURI: Multiscale Mathematical Modeling and Design Realization of Novel 2 D Functional Materials			5a. CONTRACT NUMBER W911NF-14-1-0247		
			5b. GRANT NUMBER		
			5c. PROGRAM ELEMENT NUMBER 611103		
6. AUTHORS			5d. PROJECT NUMBER		
			5e. TASK NUMBER		
			5f. WORK UNIT NUMBER		
7. PERFORMING ORGANIZATION NAMES AND ADDRESSES University of Minnesota - Minneapolis 450 McNamara Alumni Center 200 Oak Street SE Minneapolis, MN 55455 -2070			8. PERFORMING ORGANIZATION REPORT NUMBER		
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS (ES) U.S. Army Research Office P.O. Box 12211 Research Triangle Park, NC 27709-2211			10. SPONSOR/MONITOR'S ACRONYM(S) ARO		
			11. SPONSOR/MONITOR'S REPORT NUMBER(S) 65300-MA-MUR.141		
12. DISTRIBUTION AVAILABILITY STATEMENT Approved for public release; distribution is unlimited.					
13. SUPPLEMENTARY NOTES The views, opinions and/or findings contained in this report are those of the author(s) and should not be construed as an official Department of the Army position, policy or decision, unless so designated by other documentation.					
14. ABSTRACT					
15. SUBJECT TERMS					
16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT UU	15. NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON Mitchell Luskin
a. REPORT UU	b. ABSTRACT UU	c. THIS PAGE UU			19b. TELEPHONE NUMBER 612-625-6565

RPPR Final Report

as of 16-Mar-2022

Agency Code: 21XD

Proposal Number: 65300MAMUR

Agreement Number: W911NF-14-1-0247

INVESTIGATOR(S):

Name: Mitchell Luskin

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Principal: Y

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EIN: 41-6007513

Report Date: 31-Mar-2022

Date Received: 15-Mar-2022

Final Report for Period Beginning 01-Jul-2014 and Ending 31-Dec-2021

Title: MURI: Multiscale Mathematical Modeling and Design Realization of Novel 2 D Functional Materials

Begin Performance Period: 01-Jul-2014

End Performance Period: 31-Dec-2021

Report Term: 0-Other

Submitted By: Mitchell Luskin

Email: luskin@umn.edu

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Distribution Statement: 1-Approved for public release; distribution is unlimited.

STEM Degrees: 13

STEM Participants: 23

Major Goals: This MURI focused on developing mathematical tools and related computational methods for modeling the properties of two-dimensional weakly interacting multilayers in close collaboration with experimental challenges and needs. These systems became the subject of intense research efforts starting with the discovery of graphene, followed by the realization of a much broader class of 2D layered structures with semiconducting (like the transition metal di-chalcogenides) or insulating (like hexagonal boron nitride) character. More recently, the discovery of superconductivity and other correlated phases in twisted moiré systems with flat bands has our effort to investigate new moiré superlattice systems of interest and to derive and simulate models that accurately describe their electronic properties.

The novel aspect of this class of materials is the true 2D character of each layer, which interacts weakly through van der Waals forces with adjacent layers. The larger goal in the applied physics and materials communities is to combine these layers in interesting ways that will lead to devices with operational features much improved over traditional materials for optical and electronic applications. The specific goal of this MURI has been to provide the mathematical and computational underpinnings for accurately modeling these materials to guide experimental efforts toward device realization.

The MURI project focused on the development of theory and numerical methods to explore the electronic, mechanical, and diffraction properties of incommensurate 2D layered heterostructures. These methods are needed to overcome the limits of approximating incommensurate heterostructures by methods based on periodicity (supercells) and to explore the physics of incommensurate heterostructures.

The major experimental goals of this collaborative research were the characterization of 2D van der Waals heterostructures by measuring structural, electrical, thermal and mechanical properties in order provide experimental inputs and confirmation for theoretical and mathematical modeling of the team.

Accomplishments: We uploaded our accomplishments.

Training Opportunities: Mathematics, materials, engineering, and physics students and postdocs have worked closely through weekly onsite and online group meetings.

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Results Dissemination: Via presentations at conferences, seminars, and colloquia, and in papers have been uploaded.

The following are some major invited presentation based on this work:

Mitchell Luskin

Applied Math Seminar, UC Berkeley, October 7, 2014

Invited Minisymposium Lecture, Materials Research Society Meeting, Boston, December 1, 2014

Scientific Computing Seminar, Brown University, December 5, 2014

Distinguished Lecture in Applied Mathematics, University of Massachusetts, Amherst, December 8, 2014

Applied Mathematics Colloquium, Columbia University, March 10, 2015

Applied Mathematics and Mathematical Physics Colloquium, Imperial College, April 30, 2015

Invited Lecture, IPAM Arrowhead Reunion Materials Defects Workshop, June 8, 2015

Keynote Minisymposium Lecture, U.S. National Congress on Computational Mechanics, San Diego, July 27, 2015

Applied Mathematics Colloquium, MIT, November 8, 2015

Invited Lecture, Workshop on New frontiers in advanced 2D Materials (Beyond Graphene), October 15, 2015

Applied Math Colloquium, Caltech, Jan 20, 2016

Invited Lecture, IPAM Workshop on Uncertainty Quantification for Multiscale Stochastic Systems and Applications, January 22, 2016

Invited Lecture, IPAM Workshop on Partial Order: Mathematics, Simulations and Applications, January 25, 2016

Mathematics Colloquium, University of Wisconsin, Madison, March 11, 2016

Invited Lecture, Radcliffe Institute, Harvard University, April 11, 2016

Invited Lecture, PIMS Workshop on Variational Models of Fracture, Banff, May 9, 2016

Plenary speaker, 13th annual conference on "Frontiers in Applied and Computational Mathematics," NJIT, June 3--4, 2016.

Invited Lecture Series, PIRE-CNA 2016 Summer School on

New Frontiers in Nonlinear Analysis for Materials, Carnegie Mellon University, June 6, 2016

Invited Lecture, Carnegie Mellon University, Topics in Applied Nonlinear Analysis:

Recent Advances and New Trends, July 19, 2016

Invited Lecture, Banff International Research Station, Coupled Mathematical Models for Physical and Biological Nanoscale Systems and Their Applications, August 29, 2016

Invited Lecture, Center for Scientific Computation and Mathematical Modeling, University of Maryland, Sept 21, 2016

Invited Lecture, CECAM-DE-SMSM, University of Darmstadt, Germany, Multiscale Simulation Methods in Soft Matter Systems, October 4, 2016

Invited Lecture, University of Heidelberg, Oct 7, 2016

Invited Lecture, Dijon, France, Multiscale Materials Modeling (MMM) Conference, October 10, 2016

Invited Lecture, Ecole des Ponts, Paris, October 17, 2016

Le s'eminare du Laboratoire Jacques-Louis Lion, Paris, October 21, 2016

Invited Lecture, Workshop on Mathematical and Physical Aspects of Topologically Protected States, Columbia University, May 3, 2017

Keynote Lecture, U.S. National Congress on Computational Mechanics, Montreal, July 19, 2017

Invited Lecture, IMA, University of Minnesota, Aug 15, 2017

Applied Math/PDE Seminar, UCSB, Dec 1, 2017

Applied Math Seminar, UC Berkeley, March 14, 2018

Applied Math Seminar, Stanford, March 16, 2018

Invited Lecture, Fields Institute, University of Toronto, May 24, 2018

Invited Lecture, Fields Institute, University of Toronto, May 29, 2018

Invited Lecture, Euromech, University of Seville, June 10, 2018

Invited Lecture, IMA, University of Minnesota, June 28, 2018

Invited Minisymposium, SIAM Materials, July 10, 2018

Oberwolfach Lecture, October 30, 2018

Lecture Series, Newton Institute, University of Cambridge, March 23-25, 2019

Math Colloquium, UCSD, April 4, 2019

Condensed Matter Seminar, UCSD, April 9, 2019

Applied Math Seminar, University of Chicago, April, 25, 2019

Mechanics Seminar, University of Minnesota, April 30, 2019

Invited Lecture, Bernoulli Institute, EPFL, Lausanne, May 24, 2019

Invited Lecture, IPAM Lake Arrowhead, June 12, 2019

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Lecture, Oberwolfach, July 30, 2019
Invited Lecture, Institute for Theoretical Science, University of Oregon, October 30, 2019
DoD Basic Research Forum, December 19, 2019
CMX Seminar, Caltech, January 10, 2020
Invited Lecture, IPAM, January 13, 2020
Invited Lecture, AIMS/Columbia/Simons Workshop on Topological Insulators, December 10, 2020
Invited Minisymposium Lecture, SIAM CSE, March 4, 2021
Math Colloquium, University of Minnesota, March 25, 2021
Applied Math Seminar, UC Berkeley, March 31, 2021
Applied Math Seminar, Courant, April 23, 2021
SIAM Materials Science, May 17, 2021
Distinguished Lecture, Shanghai Jiao Tong University, June 1, 2021
IPAM, Lake Arrowhead, June 7, 2021
Mittag-Leffler Institute, July 12, 2021

Honors and Awards: Paul Cazeaux, MURI postdoc working with Prof. Mitchell Luskin, has won a best poster prize at the annual meeting of the French Applied Math Society (SMAI), June 8, 2015, for his poster "Cauchy-Born rule for incommensurate systems of coupled atomic chains."

Mitchell Luskin's paper "On the Wiener–Hopf Method for Surface Plasmons: Diffraction from Semiinfinite Metamaterial Sheet" with MURI collaborator Dionysius Margetis and MURI postdoc Matthias Maier, was chosen as one of the highlights of the year in the journal *Studies in Applied Math*.

MURI graduate student received the 2019 SIAM Student Paper Prize for his paper "Electronic Density of States for Incommensurate Layers," published in *Multiscale Modeling and Simulation: A SIAM Interdisciplinary Journal* in 2017. Massatt co-authored the paper with Mitchell Luskin and Christoph Ortner while he was a student at the University of Minnesota.

Philip Kim received a Vannevar Bush Faculty Fellowship in 2018.

Correlated Insulating States and Transport Signature of Superconductivity in Twisted Trilayer Graphene Superlattices by Luskin, Kaxiras, and Wang, et al. was selected to be an Editor's Suggestion in *Physical Review Letters*

Protocol Activity Status:

Technology Transfer: We invited Dr. Paul Sheehan (NRL) for seminars and discussion at Harvard. (June 16, 2015)

We collaborated with Dr. William Oliver's team in Lincoln laboratory to test out a traveling wave parametric amplifier with Josephson junctions (TWPA) for noise thermometry with high-bandwidth noise measurement techniques to extract thermal conductivity in two-dimensional systems. With a new generation of quantum-limited parametric amplifiers, we can extend our techniques to allow us to measure thermal transport on a level below the thermal conductance quantum, enabling measurements of topological edge states in van der Waals systems.

We have discussed with Dr. Madan Dubey for potential collaboration in device fabrication and exchange of samples.

We contacted Dr. Robert Younts at SPAWAR-Atlantic and Dr. Carlos Torres at NIWCr-Pacific for studying optical properties of TMD van der Waals heterostructures with a goal to fabricate ultra-sensitive optical sensors.

Stanislav Tsoi (NRL)
Stephen Hellberg (NRL)

PARTICIPANTS:

RPPR Final Report
as of 16-Mar-2022

Participant Type: PD/PI

Participant: Mitchell Luskin

Person Months Worked: 4.00

Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)

Participant: Alexander Watson

Person Months Worked: 12.00

Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Graduate Student (research assistant)

Participant: Diyi Liu

Person Months Worked: 4.00

Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)

Participant: Paul Cazeaux

Person Months Worked: 11.00

Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)

Participant: Matthias Maier

Person Months Worked: 4.00

Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)

Participant: Vera Andreeva

Person Months Worked: 11.00

Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Graduate Student (research assistant)

Participant: Daniel Massatt

Person Months Worked: 11.00

Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Co PD/PI

Participant: Philip Kim

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Person Months Worked: 1.00
Project Contribution:
National Academy Member: Y

Funding Support:

Participant Type: Graduate Student (research assistant)
Participant: Zeyu Hao

Person Months Worked: 6.00
Project Contribution:
National Academy Member: N

Funding Support:

Participant Type: Graduate Student (research assistant)
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Project Contribution:
National Academy Member: N

Funding Support:

Participant Type: Faculty
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Person Months Worked: 1.00
Project Contribution:
National Academy Member: Y

Funding Support:

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Person Months Worked: 3.00
Project Contribution:
National Academy Member: N

Funding Support:

Participant Type: Graduate Student (research assistant)
Participant: Daniel Fernandez

Person Months Worked: 1.00
Project Contribution:
National Academy Member: N

Funding Support:

Participant Type: Graduate Student (research assistant)
Participant: Elizabeth Park

Person Months Worked: 1.00
Project Contribution:
National Academy Member: N

Funding Support:

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)
Participant: Qi Yang

Person Months Worked: 12.00
Project Contribution:

Funding Support:

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National Academy Member: N

Participant Type: Graduate Student (research assistant)

Participant: Jonathan Zauberman

Person Months Worked: 6.00

Funding Support:

Project Contribution:

National Academy Member: N

Participant Type: Graduate Student (research assistant)

Participant: Wei Ren

Person Months Worked: 4.00

Funding Support:

Project Contribution:

National Academy Member: N

Participant Type: Graduate Student (research assistant)

Participant: Xi Zhang

Person Months Worked: 8.00

Funding Support:

Project Contribution:

National Academy Member: N

Participant Type: Graduate Student (research assistant)

Participant: Konstantin Davydov

Person Months Worked: 10.00

Funding Support:

Project Contribution:

National Academy Member: N

Participant Type: Graduate Student (research assistant)

Participant: Elliot Bell

Person Months Worked: 4.00

Funding Support:

Project Contribution:

National Academy Member: N

Participant Type: Co PD/PI

Participant: Efthimios Kaxiras

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Funding Support:

Project Contribution:

National Academy Member: N

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)

Participant: Mattia Angeli

Person Months Worked: 1.00

Funding Support:

Project Contribution:

National Academy Member: N

RPPR Final Report
as of 16-Mar-2022

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)

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Person Months Worked: 3.00

Funding Support:

Project Contribution:

National Academy Member: N

Participant Type: Graduate Student (research assistant)

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Person Months Worked: 3.00

Funding Support:

Project Contribution:

National Academy Member: N

Participant Type: Co PD/PI

Participant: Ellad Tadmor

Person Months Worked: 1.00

Funding Support:

Project Contribution:

National Academy Member: N

Participant Type: Co PD/PI

Participant: Petr Plechac

Person Months Worked: 1.00

Funding Support:

Project Contribution:

National Academy Member: N

Participant Type: Faculty

Participant: Ke Wang

Person Months Worked: 1.00

Funding Support:

Project Contribution:

National Academy Member: N

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)

Participant: Kapildep Dolui

Person Months Worked: 1.00

Funding Support:

Project Contribution:

National Academy Member: N

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)

Participant: Marko Petrovic

Person Months Worked: 2.00

Funding Support:

Project Contribution:

National Academy Member: N

RPPR Final Report
as of 16-Mar-2022

Participant Type: Graduate Student (research assistant)
Participant: Gabriel Song
Person Months Worked: 1.00
Project Contribution:
National Academy Member: N
Funding Support:

Participant Type: Graduate Student (research assistant)
Participant: Kan-Ting Tsai
Person Months Worked: 7.00
Project Contribution:
National Academy Member: N
Funding Support:

Participant Type: Graduate Student (research assistant)
Participant: Xi Zhang
Person Months Worked: 7.00
Project Contribution:
National Academy Member: N
Funding Support:

Participant Type: Graduate Student (research assistant)
Participant: Stephen Carr
Person Months Worked: 1.00
Project Contribution:
National Academy Member: N
Funding Support:

ARTICLES:

Publication Type: Journal Article
Journal: Physical Review Letters
Publication Identifier Type: DOI
Volume: 1.15E+002 Issue: 1.0E+000 First Page #: 0
Date Submitted:
Publication Location:
Peer Reviewed: Y
Publication Status: 1-Published
Publication Identifier: 10.1103/PhysRevLett.115.017002
Date Published:

Article Title: Revealing the Empty-State Electronic Structure of Single-Unit-Cell FeSe/SrTiO₃

Authors:

Keywords: Pnictides and chalcogenides, Electronic structure, Tunneling phenomena: single particle tunneling and STM, Superconducting films and low-dimensional structures

Abstract: We use scanning tunneling spectroscopy to investigate the filled and empty electronic states of superconducting single-unit-cell FeSe deposited on SrTiO₃(001). We map the momentum-space band structure by combining quasiparticle interference imaging with decay length spectroscopy. In addition to quantifying the filled-state bands, we discover a Γ -centered electron pocket 75 meV above the Fermi energy. Our density functional theory calculations show the orbital nature of empty states at Γ and explain how the Se height is a key tuning parameter of their energies, with broad implications for electronic properties.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors
Acknowledged Federal Support:

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Applied Mathematics Research Express

Publication Identifier Type: DOI

Publication Identifier: dx.doi.org/10.1093/amrx/abv007

Volume: 2

Issue: 1

First Page #: 329

Date Submitted: 8/23/17 12:00AM

Date Published: 8/5/15 4:00AM

Publication Location:

Article Title: Computational error estimates for Born-Oppenheimer molecular dynamics with nearly crossing potential surfaces

Authors: C. Bayer, H. Hoel, A. Kadir, P. Plechac, M. Sandberg, A. Szepessy

Keywords: Schrodinger equation, Born-Oppenheimer approximation, ab-initio molecular dynamics, semiclassical approximations, Weyl calculus

Abstract: The difference of the values of observables for the time-independent Schrödinger equation, with matrix valued potentials, and the values of observables for $\{\text{it ab initio}\}$ Born-Oppenheimer molecular dynamics, on the ground state, depends on the probability to be in excited states and the electron/nuclei mass ratio. The paper first proves an error estimate (depending on the electron/nuclei mass ratio and the probability to be in excited states) for this difference of microcanonical observables, assuming that molecular dynamic space-time averages converge, with a rate related to the maximal Lyapunov exponent. The error estimate is uniform in the number of particles and the analysis does not assume a uniform lower bound on the spectral gap of the electron operator and consequently the probability to be in excited states can be large. A numerical method to determine the probability to be in excited states is then presented, based on Ehrenfest molecular dynamics and stability analysis.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: PHYSICAL REVIEW B (Submitted)

Publication Identifier Type: DOI

Publication Identifier: 10.1103/PhysRevB.92.205108

Volume: 92

Issue: 0

First Page #: 205108

Date Submitted: 8/8/18 12:00AM

Date Published: 11/5/15 10:00AM

Publication Location:

Article Title: Ab-initio Tight-Binding Hamiltonian for Transition Metal Dichalcogenides

Authors: Fang, S.; Defo, R. K.; Shirodkar, S. N.; Lieu, S.; Tritsarlis, G.A.; Kaxiras, E.

Keywords: Methods of electronic structure calculations, Electronic structure of nanoscale materials and related systems, Multilayers, superlattices, heterostructures, Other inorganic semiconductors

Abstract: We present an accurate ab-initio tight-binding hamiltonian for the transition-metal dichalcogenides, MoS₂, MoSe₂, WS₂, WSe₂, with a minimal basis (the d orbitals for the metal atoms and p-orbitals for the chalcogen atoms) based on a transformation of the Kohn-Sham density function theory (DFT) hamiltonian to a basis of maximally localized Wannier functions (MLWF). The truncated tight-binding hamiltonian (TBH), with only on-site, first and partial second neighbor interactions, including spin-orbit coupling, provides a simple physical picture and the symmetry of the main band-structure features. Interlayer interactions between adjacent layers are modeled by transferable hopping terms between the chalcogen p orbitals. The full-range tight-binding hamiltonian (FTBH) can be reduced to hybrid-orbital k.p effective hamiltonians near the band extrema that captures important low-energy excitations. These ab-initio hamiltonians can serve as the starting point for applications to interacting materials.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: 2D MATERIALS

Publication Identifier Type: DOI

Publication Identifier: 10.1088/2053-1583/3/1/015005

Volume: 3

Issue: 1

First Page #: 0

Date Submitted: 8/18/18 12:00AM

Date Published: 1/27/16 3:00PM

Publication Location:

Article Title: Excitation intensity dependence of photoluminescence from monolayers of MoS2 and MoS2/WS2 heterostructures

Authors: Kaplan, D.; Gong, Y.; Mills, K.; Swaminathan, V.; Ajayan, P.M.; Shirodkar, S.; Kaxiras, E.

Keywords: excitons, photoluminescence, monolayers, metal dichalcogenides

Abstract: A detailed study of the excitation dependence of the photoluminescence from monolayers of MoS2 and MoS2/WS2 heterostructures grown by chemical vapor deposition on Si substrates has revealed that the luminescence from band edge excitons from MoS2 monolayers shows a linear dependence on excitation intensity for both above band gap and resonant excitation conditions. In particular, a band separated by ~ 55 meV from the A exciton, referred to as the C band, shows the same linear dependence on excitation intensity as the band edge excitons. A band similar to the C band has been previously ascribed to a trion, a charged three-particle exciton. However, in our study the C band does not show the 3/2 power dependence on excitation intensity as would be expected for a three particle exciton. Further, the photoluminescence from the MoS2 monolayer in a bilayer MoS2/WS2 hetero-stack, under resonant excitation conditions where only the MoS2 absorbs the laser energy, also revealed a linear dependence

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: PHYSICAL REVIEW B

Publication Identifier Type: DOI

Publication Identifier: doi.org/10.1103/PhysRevB.94.155310

Volume: 94

Issue: 0

First Page #: 155310

Date Submitted: 8/30/17 12:00AM

Date Published: 10/27/16 4:00AM

Publication Location:

Article Title: Strain Dependence of Structural and Electronic Properties of Select Two-Dimensional Transition Metal Dichalcogenides: A First-Principles Analysis

Authors: R. K. Deho, S. Fang, S. N. Shirodkar, G. A. Tritsarlis, A. Dimoulas, and E. Kaxiras

Keywords: monolayers, 2D materials, transition metal dichalcogenides, electronic properties

Abstract: As a result of the extensive work done on graphene, leading to advances in sample preparation, optical detection, and transfer and manipulation of two-dimensional (2D) materials, there has been a resurgence of interest in layered materials from which single sheets can be extracted. Like graphene, transition metal dichalcogenides (TMDCs) can be exfoliated as single sheets, but, unlike graphene, certain TMDCs exhibit a non-zero band gap which can be used to achieve high on/off ratios in transistor circuits and other applications. To understand emergent properties of compounds consisting of TMDCs layered with other materials where there is a lattice mismatch, it is crucial to understand the dependence of the key structural and electronic properties on strain. To this end, we present a phase diagram of the transition from an indirect to a direct band gap as a function of strain for the conventional MoS2, MoSe2 and WS2, WSe2 compounds, and predict the emergent properties of the hybrid MoSS

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Journal of Applied Physics

Publication Identifier Type: DOI

Publication Identifier: 10.1063/1.4939616

Volume: 119

Issue: 2

First Page #: 025301

Date Submitted: 6/28/16 12:00AM

Date Published: 1/2/16 11:00AM

Publication Location:

Article Title: Manipulation of surface plasmon polariton propagation on isotropic and anisotropic two-dimensional materials coupled to boron nitride heterostructures

Authors: Sandeep Inampudi, Mina Nazari, Ali Forouzmand, Hossein Mosallaei

Keywords: plasmonics, 2D sheets

Abstract: We present a comprehensive analysis of surface plasmon polariton dispersion characteristics associated with isotropic and anisotropic two-dimensional atomically thin layered materials (2D sheets) coupled to h-BN heterostructures. A scattering matrix based approach is presented to compute the electromagnetic fields and related dispersion characteristics of stacked layered systems composed of anisotropic 2D sheets and uniaxial bulk materials. We analyze specifically the surface plasmon polariton (SPP) dispersion characteristics in case of isolated and coupled two-dimensional layers with isotropic and anisotropic conductivities. An analysis based on residue theorem is utilized to identify optimum optical parameters (surface conductivity) and geometrical parameters (separation between layers) to maximize the SPP field at a given position. The effect of type and degree of anisotropy on the shapes of iso-frequency curves and propagation characteristics is discussed in detail. The analysis presents

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Applied Optics

Publication Identifier Type: DOI

Publication Identifier: 10.1364/AO.55.002967

Volume: 55

Issue: 11

First Page #: 2967

Date Submitted: 6/28/16 12:00AM

Date Published: 4/1/16 8:00AM

Publication Location:

Article Title: Light manipulation with flat and conformal inhomogeneous dispersive impedance sheets: an efficient FDTD modeling

Authors: Samad Jafar-Zanjani, Jierong Cheng, Hossein Mosallaei

Keywords: plasmonics, 2D sheets, modeling

Abstract: An efficient auxiliary differential equation method for incorporating 2D inhomogeneous dispersive impedance sheets in the finite-difference time-domain solver is presented. This unique proposed method can successfully solve optical problems of current interest involving 2D sheets. It eliminates the need for ultrafine meshing in the thickness direction, resulting in a significant reduction of computation time and memory requirements. We apply the method to characterize a novel broad-beam leaky-wave antenna created by cascading three sinusoidally modulated reactance surfaces and also to study the effect of curvature on the radiation characteristic of a conformal impedance sheet holographic antenna. Considerable improvement in the simulation time based on our technique in comparison with the traditional volumetric model is reported. Both applications are of great interest in the field of antennas and 2D sheets.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Journal of Mathematical Physics

Publication Identifier Type: DOI

Publication Identifier: 10.1063/1.4945083

Volume: 57

Issue: 4

First Page #: 042903

Date Submitted: 6/28/16 12:00AM

Date Published: 4/1/16 5:00AM

Publication Location:

Article Title: On solutions of Maxwell's equations with dipole sources over a thin conducting film

Authors: Dionisios Margetis, Mitchell Luskin

Keywords: graphene, surface plasmon-polariton

Abstract: We derive and interpret solutions of time-harmonic Maxwell's equations with a vertical and a horizontal electric dipole near a planar, thin conducting film, e.g., graphene sheet, lying between two unbounded isotropic and non-magnetic media. Exact expressions for all field components are extracted in terms of rapidly convergent series of known transcendental functions when the ambient media have equal permittivities and both the dipole and observation point lie on the plane of the film. These solutions are simplified for all distances from the source when the film surface resistivity is large in magnitude compared to the intrinsic impedance of the ambient space. The formulas reveal the analytical structure of two types of waves that can possibly be excited by the dipoles and propagate on the film. One of these waves is intimately related to the surface plasmon-polariton of transverse-magnetic polarization of plane waves.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Journal of Materials Research

Publication Identifier Type: DOI

Publication Identifier: 10.1557/jmr.2016.99

Volume: 31

Issue: 07

First Page #: 959

Date Submitted: 6/28/16 12:00AM

Date Published: 3/1/16 6:00AM

Publication Location:

Article Title: Perturbation theory for weakly coupled two-dimensional layers

Authors: Georgios A. Tritsarlis, Sharmila N. Shirodkar, Efthimios Kaxiras, Paul Cazeaux, Mitchell Luskin, Petr Ple

Keywords: electronic structure, two-dimensional layers

Abstract: A key issue in two-dimensional structures composed of atom-thick sheets of electronic materials is the dependence of the properties of the combined system on the features of its parts. Here, we introduce a simple framework for the study of the electronic structure of layered assemblies based on perturbation theory. Within this framework, we calculate the band structure of commensurate and twisted bilayers of graphene (Gr) and hexagonal boron nitride (h-BN), and of a Gr/h-BN heterostructure, which we compare with reference full-scale density functional theory calculations. This study presents a general methodology for computationally efficient calculations of twodimensional materials and also demonstrates that for relatively large twist in the Gr bilayer, the perturbation of electronic states near the Fermi level is negligible.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

RPPR Final Report
as of 16-Mar-2022

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: J. Comput. Phys.
Publication Identifier Type: DOI **Publication Identifier:** doi.org/10.1016/j.jcp.2017.03.014
Volume: 339 **Issue:** **First Page #:** 126
Date Submitted: 8/5/17 12:00AM **Date Published:** 6/15/17 10:05PM
Publication Location:

Article Title: Dipole excitation of surface plasmon on a conducting sheet: finite element approximation and validation

Authors: MATTHIAS MAIER^L, DIONISIOS MARGETIS, AND MITCHELL LUSKIN

Keywords: Time-harmonic Maxwell's equations, finite element method, surface plasmonpolariton, weak discontinuity on hypersurface

Abstract: By using a suitable rescaled form of time-harmonic Maxwell's equations, we numerically simulate the propagation of a slowly decaying electromagnetic wave, called surface plasmonpolariton, excited along a large conducting sheet, e.g., a single-layer graphene sheet, by an electric Hertzian dipole. For this purpose, we derive a variational formulation that enables a direct numerical treatment of the associated class of boundary value problems by finite elements. The conducting sheet is modeled as an idealized hypersurface with an effective electric conductivity. The requisite weak discontinuity for the magnetic field across the hypersurface can be incorporated naturally into the variational formulation. We carry out numerical simulations for an infinite sheet with constant isotropic conductivity embedded in two spatial dimensions; and test our numerics against the closed-form exact solution obtained by the Fourier transform in the tangential coordinate. Numerical aspects of our

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors
Acknowledged Federal Support: Y

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: SIAM Journal of Multiscale Modeling and Simulation
Publication Identifier Type: DOI **Publication Identifier:** 10.1137/16M1076198
Volume: 15 **Issue:** 1 **First Page #:** 56
Date Submitted: 8/5/17 12:00AM **Date Published:** 4/13/17 10:17PM
Publication Location:

Article Title: ANALYSIS OF RIPPLING IN INCOMMENSURATE ONE-DIMENSIONAL COUPLED CHAINS

Authors: PAUL CAZEAUX^L, MITCHELL LUSKIN^L, AND ELLAD B. TADMOR

Keywords: 2D materials, heterostructures, ripples, incommensurability, atomistic relaxation, Frenkel-Kontorova

Abstract: Graphene and other recently developed 2D materials exhibit exceptionally strong in-plane stiffness. Relaxation of few-layer structures, either free-standing or on slightly mismatched substrates occurs mostly through out-of-plane bending and the creation of large-scale ripples. In this work, we present a novel double chain model, where we allow relaxation to occur by bending of the incommensurate coupled system of chains. As we will see, this model can be seen as a new application of the well-known Frenkel-Kontorova model for a one-dimensional atomic chain lying in a periodic potential. We focus in particular on modeling and analyzing ripples occurring in ground state configurations, as well as their numerical simulation.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors
Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Modeling and Simulations in Materials Science and Engineering

Publication Identifier Type: Other

Publication Identifier:

Volume:

Issue:

First Page #:

Date Submitted: 7/5/16 12:00AM

Date Published: 6/29/16 6:21PM

Publication Location:

Article Title: A KIM-compliant potfit for fitting sloppy interatomic potentials: Application to the EDIP model for silicon

Authors: Mingjian Wen, Junhao Li, Peter Brommer, Ryan S. Elliott, James P. Sethna and Ellad B. Tadmor

Keywords: Interatomic potentials; force matching; geodesic Levenberg-Marquardt

Abstract: The potfit program is an implementation of the force-matching method for fitting interatomic potentials. We extended potfit to be compliant with the KIM Application Programming Interface (API) standard (part of the Knowledgebase of Interatomic Models Project) and incorporated a geodesic Levenberg-Marquardt (LM) minimization algorithm. The extended potfit code is the first step in developing a KIM-based fitting framework for interatomic potentials for bulk and two-dimensional materials. The code is available for download via <https://www.potfit.net>.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

Publication Type: Journal Article

Peer Reviewed: Y

Publication Status: 1-Published

Journal: Physical Review B

Publication Identifier Type: DOI

Publication Identifier: 10.1103/PhysRevB.93.235153

Volume: 93

Issue: 23

First Page #:

Date Submitted: 6/29/16 12:00AM

Date Published: 6/1/16 4:00AM

Publication Location:

Article Title: Electronic structure theory of weakly interacting bilayers

Authors: Shiang Fang, Efthimios Kaxiras

Keywords: two-dimensional materials

Abstract: We derive electronic structure models for weakly interacting bilayers such as graphene-graphene and graphene-hexagonal boron nitride, based on density functional theory calculations followed by Wannier transformation of electronic states. These transferable interlayer coupling models can be applied to investigate the physics of bilayers with arbitrary translations and twists. The functional form, in addition to the dependence on the distance, includes the angular dependence that results from higher angular momentum components in the Wannier pz orbitals. We demonstrate the capabilities of the method by applying it to a rotated graphene bilayer, which produces the analytically predicted renormalization of the Fermi velocity, Van Hove singularities in the density of states, and moiré pattern of the electronic localization at small twist angles. We further extend the theory to obtain the effective couplings by integrating out neighboring layers. This approach is instrumental for the design

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

RPPR Final Report
as of 16-Mar-2022

Publication Type: Journal Article

Peer Reviewed: Y

Publication Status: 1-Published

Journal: Science

Publication Identifier Type: DOI

Publication Identifier: 10.1126/science.aad0343

Volume: 351

Issue: 6277

First Page #: 1058

Date Submitted: 7/5/16 12:00AM

Date Published: 2/1/16 12:00AM

Publication Location:

Article Title: Observation of the Dirac fluid and the breakdown of the Wiedemann-Franz law in graphene

Authors: J. Crossno, J. K. Shi, K. Wang, X. Liu, A. Harzheim, A. Lucas, S. Sachdev, P. Kim, T. Taniguchi, K. Wat

Keywords: Dirac fluid, Wiedemann-Franz law, graphene

Abstract: Interactions between particles in quantum many-body systems can lead to collective behavior described by hydrodynamics. One such system is the electron-hole plasma in graphene near the charge neutrality point, which can form a strongly coupled Dirac fluid. This charge neutral plasma of quasi-relativistic fermions is expected to exhibit a substantial enhancement of the thermal conductivity, thanks to decoupling of charge and heat currents within hydrodynamics. Employing high sensitivity Johnson noise thermometry, we report an order of magnitude increase in the thermal conductivity and the breakdown of the Wiedemann-Franz law in the thermally populated charge neutral plasma in graphene. This result is a signature of the Dirac fluid, and constitutes direct evidence of collective motion in a quantum electronic fluid.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

Publication Type: Journal Article

Peer Reviewed: Y

Publication Status: 1-Published

Journal: Mathematical modeling and numerical analysis

Publication Identifier Type: DOI

Publication Identifier: doi.org/10.1051/m2an/2017057

Volume: 52

Issue:

First Page #: 729

Date Submitted: 8/9/18 12:00AM

Date Published: 7/6/16 6:29AM

Publication Location:

Article Title: Cauchy-Born strain energy density for coupled incommensurate elastic chains

Authors: Paul Cazeaux and Mitchell Luskin

Keywords: Cauchy-Born strain energy density, coupled incommensurate elastic chains

Abstract: The recent fabrication of weakly interacting incommensurate two-dimensional lattices \cite{geimvdw} requires an extension of the classical notion of the Cauchy-Born strain energy density since these atomistic systems are not periodic. In this paper, we rigorously formulate and analyze a Cauchy-Born strain energy density for weakly interacting incommensurate one-dimensional lattices (chains) as a large body limit and we give error estimates for its approximation by the popular supercell method.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Physical Review B

Publication Identifier Type: DOI

Publication Identifier: 10.1103/PhysRevB.93.245438

Volume: 93

Issue: 24

First Page #:

Date Submitted: 8/1/16 12:00AM

Date Published: 6/1/16 1:00PM

Publication Location: College Park, MD

Article Title: Li intercalation at graphene/hexagonal boron nitride interfaces

Authors: Sharmila N. Shirodkar, Efthimios Kaxiras

Keywords: Intercalation, graphene/hexagonal boron nitride interfaces

Abstract: Intercalation of Li in graphite and other layered structures is of interest for highly efficient energy storage devices. In this paper, we determine the extent to which Li intercalates at the different interfaces formed between graphene (G) and hexagonal boron nitride (hBN) heterostructures. We use ab initio calculations to explore in detail the position of the dispersed Li atoms, changes in the structure at the interfaces, energetic stability of the configurations, and the corresponding electronic structure with varying concentrations of the intercalant. We trace the origin of the energetic stability and maximum concentration of Li that intercalates into various layered structures to the ability of the interface to accept electrons. Our calculations indicate that Li intercalates easiest at G/G interfaces, followed by interfaces between G/hBN, whereas Li cannot intercalate in hBN/hBN interfaces. Our results provide a framework for the design of experimental setups with optimal Li inter

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Physical Review B

Publication Identifier Type: DOI

Publication Identifier: 10.1103/PhysRevB.93.125129

Volume: 93

Issue: 12

First Page #:

Date Submitted: 8/1/16 12:00AM

Date Published: 3/1/16 6:00AM

Publication Location:

Article Title: Bounds on nanoscale nematicity in single-layer FeSe/SrTiO₃

Authors: Dennis Huang, Tatiana A. Webb, Shiang Fang, Can-Li Song, Cui-Zu Chang, Jagadeesh S. Moodera, Ef

Keywords: nanoscale nematicity, single-layer FeSe/SrTiO₃

Abstract: We use scanning tunneling microscopy (STM) and quasiparticle interference (QPI) imaging to investigate the low-energy orbital texture of single-layer FeSe/SrTiO₃. We develop a T-matrix model of multiorbital QPI to disentangle scattering intensities from Fe 3dxz and 3dyz bands, enabling the use of STM as a nanoscale detection tool of nematicity. By sampling multiple spatial regions of a single-layer FeSe/SrTiO₃ film, we quantitatively exclude static xz/yz orbital ordering with domain size larger than $\ell^2 = 20 \text{ nm} \times 20 \text{ nm}$, xz/yz Fermi wave vector difference larger than $\Delta k = 0.014$, and energy splitting larger than $\Delta E = 3.5 \text{ meV}$. The lack of detectable ordering pinned around defects places qualitative constraints on models of fluctuating nematicity.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: SIAM Multiscale Modeling and Simulation

Publication Identifier Type: DOI

Publication Identifier: 10.1137/16M1088363

Volume: 15

Issue: 1

First Page #: 476

Date Submitted: 8/5/17 12:00AM

Date Published: 5/19/17 5:40AM

Publication Location:

Article Title: Electronic Density of States for Incommensurate Layers

Authors: Daniel Massatt, Mitchell Luskin, Christoph Ortner

Keywords: Electronic Density of States, 2D, Incommensurate

Abstract: We prove that the electronic density of states (DOS) for 2D incommensurate layered structures, where Bloch theory does not apply, is well-defined as the thermodynamic limit of finite clusters. In addition, we obtain an explicit representation formula for the DOS as an integral over local configurations. Next, based on this representation formula, we propose a novel algorithm for computing electronic structure properties in incommensurate heterostructures, which overcomes limitations of the common approach to artificially strain a large supercell and then apply Bloch theory.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

Publication Type: Journal Article

Peer Reviewed: Y

Publication Status: 1-Published

Journal: Applied Optics

Publication Identifier Type: DOI

Publication Identifier: 10.1364/AO.56.003132

Volume: 56

Issue: 11

First Page #: 3132

Date Submitted: 8/9/17 12:00AM

Date Published: 4/1/17 2:00PM

Publication Location:

Article Title: Graphene-based near-field optical microscopy: high-resolution imaging using reconfigurable gratings

Authors: Sandeep Inampudi, Jierong Cheng, Hossein Mosallaei

Keywords: nanomaterials, diffraction grating

Abstract: High-resolution and fast-paced optical microscopy is a requirement for current trends in biotechnology and materials industry. The most reliable and adaptable technique so far to obtain higher resolution than conventional microscopy is near-field scanning optical microscopy (NSOM), which suffers from a slow-paced nature. Stemming from the principles of diffraction imaging, we present fast-paced graphene-based scanning-free widefield optical microscopy that provides image resolution that competes with NSOM. Instead of spatial scanning of a sharp tip, we utilize the active reconfigurable nature of graphene's surface conductivity to vary the diffraction properties of a planar digitized atomically thin graphene sheet placed in the near field of an object. Scattered light through various realizations of gratings is collected at the far-field distance and postprocessed using a transmission function of surface gratings developed on the principles of rigorous coupled wave analysis.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: ACS Omega

Publication Identifier Type: DOI

Publication Identifier: 10.1021/acsomega.6b00310

Volume: 1

Issue: 5

First Page #: 843

Date Submitted: 8/9/17 12:00AM

Date Published: 11/1/16 2:00PM

Publication Location:

Article Title: Fresnel Refraction and Diffraction of Surface Plasmon Polaritons in Two-Dimensional Conducting Sheets

Authors: Sandeep Inampudi, Hossein Mosallaei

Keywords: graphene, SPP, Fresnel law

Abstract: The propagation of surface plasmon polaritons (SPPs) along two-dimensional (2D) materials, such as graphene, is a complex phenomenon linking the microscale electronic properties to macroscale optical properties. Complex geometries increase the complexity of understanding the nature and performance of optoelectronic devices based on surface wave propagation. Here, we demonstrate that under a proper design of macroscopic conductivity profile, the propagation characteristics of SPPs in 2D materials can be made analogous to the propagation of plane waves in homogeneous layers with minimal out-ofplane scattering. Such a direct resemblance enables prediction, design, and calculation of SPP propagation through advanced geometries using fundamental laws of optics. We demonstrate that the propagation of surface waves can be manipulated in-plane using reflection, refraction, diffraction, and also generalized refraction laws analogous to plane waves.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Physical Review B

Publication Identifier Type: DOI

Publication Identifier: 10.1103/PhysRevB.94.235403

Volume: 94

Issue: 23

First Page #:

Date Submitted: 8/9/17 12:00AM

Date Published: 12/1/16 4:00PM

Publication Location:

Article Title: Mechanical actuation of graphene sheets via optically induced forces

Authors: Mohammad Mahdi Salary, Sandeep Inampudi, Kuan Zhang, Ellad B. Tadmor, Hossein Mosallaei

Keywords: graphene, optical forces

Abstract: In this paper, we theoretically demonstrate the strong mechanical response of graphene sheets actuated by near-field optical forces. We study single-layer graphene and a two-layer graphene stack with large separation and show that tunable attractive and repulsive forces can be generated. A large nonlinear mechanical response can be obtained by driving the sheets through external radiation and guided modes. We report formation of graphene bubbles of several nanometers in height. Our study points towards new routes for mechanical actuation of graphene, providing new platforms for straintronics and flexible optoelectronics.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Physical Review B

Publication Identifier Type: DOI

Publication Identifier: 10.1103/PhysRevB.95.075420

Volume: 95

Issue: 7

First Page #:

Date Submitted: 8/5/17 12:00AM

Date Published: 2/1/17 6:00AM

Publication Location:

Article Title: Twistronics: Manipulating the electronic properties of two-dimensional layered structures through their twist angle

Authors: Stephen Carr, Daniel Massatt, Shiang Fang, Paul Cazeaux, Mitchell Luskin, Efthimios Kaxiras

Keywords: local density of states, two-dimensional layered structures

Abstract: The ability in experiments to control the relative twist angle between successive layers in two-dimensional (2D) materials offers an approach to manipulating their electronic properties; we refer to this approach as “twistronics.” A major challenge to theory is that, for arbitrary twist angles, the resulting structure involves incommensurate (aperiodic) 2D lattices. Here, we present a general method for the calculation of the electronic density of states of aperiodic 2D layered materials, using parameter-free Hamiltonians derived from ab initio density-functional theory. We use graphene, a semimetal, and MoS₂, a representative of the transition-metal dichalcogenide family of 2D semiconductors, to illustrate the application of our method, which enables fast and efficient simulation of multilayered stacks in the presence of local disorder and external fields. We comment on the interesting features of their density of states as a function of twist angle and local configuration and on h

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Journal of Mathematical Physics

Publication Identifier Type: DOI

Publication Identifier: 10.1063/1.4984041

Volume: 58

Issue: 6

First Page #: 063502

Date Submitted: 9/19/17 12:00AM

Date Published: 6/1/17 7:00PM

Publication Location:

Article Title: Generalized Kubo formulas for the transport properties of incommensurate 2D atomic heterostructures

Authors: Eric Cancès, Paul Cazeaux, Mitchell Luskin

Keywords: Kubo, incommensurate

Abstract: We give an exact formulation for the transport coefficients of incommensurate twodimensional atomic multilayer systems in the tight-binding approximation. This formulation is based upon the C^* -algebra framework introduced by Bellissard and collaborators [Coherent and Dissipative Transport in Aperiodic Solids, Lecture Notes in Physics (Springer, 2003), Vol. 597, pp. 413–486 and J. Math. Phys. 35(10), 5373–5451 (1994)] to study aperiodic solids (disordered crystals, quasicrystals, and amorphous materials), notably in the presence of magnetic fields (quantum Hall effect). We also present numerical approximations and test our methods on a one-dimensional incommensurate bilayer system.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Journal of Computational and Theoretical Nanoscience

Publication Identifier Type: DOI

Publication Identifier: doi:10.1166/jctn.2017.6807

Volume: 14

Issue: 7

First Page #: 3359

Date Submitted: 8/9/17 12:00AM

Date Published: 7/31/17 7:00AM

Publication Location:

Article Title: An Integral Equation Based Computational Model for Simulating Substrate-Supported 2D Sheet Structures

Authors: Shifei Tao, Samad Jafar-Zanjani, Sandeep Inampudi, and Hossein Mosallaei

Keywords: Numerical Analysis, Integral Equation (IE)-Based Methods, Domain Decomposition Method (DDM), Metamaterials, Two-Dimensional (2-D) Materials, Graphene, Surface Waves

Abstract: In this paper, we propose an integral equation (IE)-based computational model for electromagnetic (EM) characterization of freestanding and substrate-supported 2D sheet structures. The freestanding 2D sheet is modeled with surface integral equation (SIE) method by combining the impedance boundary condition (IBC), while regarding the thickness of 2D sheet as zero. For the substrate supported 2D sheet structure, we start from the coupled volume and surface integral equations (VSIE) based on the field equivalence principle. Because of the infinitesimal thickness of 2D sheets and the introduction of the equivalent surface currents on the sheet, the volume integral has been successfully transformed to a surface integral. The domain decomposition method (DDM) is also employed in which decomposes the original large problem into smaller discrete sub-domain problems and brings the reduction both in computational time and memory

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

Publication Type: Journal Article Peer Reviewed: N **Publication Status:** 5-Submitted

Journal: preprint arxiv: 1611.09784v1, submitted

Publication Identifier Type:

Publication Identifier:

Volume:

Issue:

First Page #:

Date Submitted: 8/23/17 12:00AM

Date Published:

Publication Location:

Article Title: Multi-level Monte Carlo acceleration of computations on multi-layer materials with random defects

Authors: P. Plechac, E. von Schwerin

Keywords: multi-level Monte Carlo, random defects, density of states

Abstract: We propose a Multi-level Monte Carlo technique to accelerate Monte Carlo sampling for approximation of properties of materials with random defects. The computational efficiency is investigated on test problems given by tight-binding models of a single layer of graphene or of MoS₂ where the integrated electron density of states per unit area is taken as a representative quantity of interest. For the chosen test problems the multi-level Monte Carlo estimators significantly reduce the computational time of standard Monte Carlo estimators to obtain a given accuracy.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

RPPR Final Report

as of 16-Mar-2022

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Extreme Mechanics Letters

Publication Identifier Type: DOI

Publication Identifier: 10.1016/j.eml.2016.10.010

Volume: 14

Issue:

First Page #: 16

Date Submitted: 8/24/17 12:00AM

Date Published: 11/2/16 5:08PM

Publication Location:

Article Title: Energy and moire? patterns in 2D bilayers in translation and rotation: A study using an efficient discrete-continuum interlayer potential

Authors: Kuan Zhang, Ellad B. Tadmor

Keywords: Graphene, Van der Waals, Interlayer energy, Moire? pattern

Abstract: Layered 2D materials consist of a stack of 2D materials interacting weakly via van der Waals (vdW) forces. The vdW interactions are modeled using long-range interlayer potentials, such as Lennard-Jones (LJ) and Kolmogorov-Crespi (KC), that are expensive to compute in atomistic and multiscale simulations. We propose a discrete-continuum (DC) effective potential for LJ and KC that includes full atomistic resolution in the short range and a continuum integral approximation outside. This reduces the computational cost by two orders of magnitude while retaining excellent accuracy. With this method, we examine the relation between stacking states and interlayer energy of a graphene bilayer under relative in-plane translations and rotations. We compare the performance of LJ and KC and characterize the moire? patterns under rotation. We find that the energy of an infinite graphene bilayer is constant with rotation (with discontinuities at special angles) and explain this in terms of the moire?

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 5-Submitted

Journal: Physical Review Letters

Publication Identifier Type:

Publication Identifier:

Volume:

Issue:

First Page #:

Date Submitted: 8/29/17 12:00AM

Date Published:

Publication Location:

Article Title: Structural and electron diffraction scaling of twisted graphene bilayers

Authors: Kuan Zhang, Ellad Tadmor

Keywords: Van der Waals heterostructures, graphene, electron diffraction, multiscale modeling

Abstract: A novel continuum-atomistic framework is used to simulate twisted graphene bilayers. The results show a transformation to a commensurate microstructure consisting of quasiperiodic cells containing alternating low energy AB and BA domains separated by SP solitons surrounding a high-energy AA domain. During the transformation, the size of the AA domains is minimized through additional uniform local twisting, centered on the AA domains, which scales with the initial rotation between the layers. Two regimes are observed. For small initial rotation, the twist angle is constant; for large initial rotation this value scales linearly with the applied rotation. The relaxation at small initial rotation is associated with a novel electron diffraction pattern involving the appearance of weak satellite peaks, which is explained by which is explained by a simple kinematic Gaussian rotation model. Both the angle-dependent scaling and diffraction patterns are in quantitative agreement with experiment

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Journal of Applied Physics

Publication Identifier Type: DOI

Publication Identifier: 10.1063/1.5007842

Volume: 122

Issue:

First Page #: 244301

Date Submitted: 8/17/20 12:00AM

Date Published: 12/21/17 4:00PM

Publication Location:

Article Title: A force-matching Stillinger-Weber potential for MoS₂: Parameterization and Fisher information theory based sensitivity analysis

Authors: Mingjian Wen, Sharmilla Shirodkar, Petr Plechac, Efthimios Kaxiras, Ryan Elliott, Ellad Tadmor

Keywords: potential energy surfaces, 2-dimensional systems, Density functional theory, molecular dynamics

Abstract: Two-dimensional molybdenum disulfide (MoS₂) is a promising material for the next generation of switchable transistors and photodetectors. In order to perform large-scale molecular simulations of the mechanical and thermal behavior of MoS₂-based devices, an accurate interatomic potential is required. To this end, we have developed a Stillinger-Weber potential for monolayer MoS₂. The potential parameters are optimized to reproduce the geometry (bond lengths and bond angles) of MoS₂ in its equilibrium state and to match as closely as possible the forces acting on the atoms along a dynamical trajectory obtained from ab initio molecular dynamics.

Verification calculations indicate that the new potential accurately predicts important material properties including the strain dependence of the cohesive energy, the elastic constants, and the linear thermal expansion coefficient. The uncertainty in the potential parameters is determined using a Fisher information theory analysis.

Distribution Statement: 2-Distribution Limited to U.S. Government agencies only; report contains proprietary info
Acknowledged Federal Support: Y

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Proceedings of the National Academy of Sciences

Publication Identifier Type: DOI

Publication Identifier: 10.1073/pnas.1610204114

Volume: 114

Issue: 2

First Page #: 263

Date Submitted: 8/18/18 12:00AM

Date Published: 1/1/17 8:00PM

Publication Location:

Article Title: Relationship between local structure and relaxation in out-of-equilibrium glassy systems

Authors: Samuel S. Schoenholz, Ekin D. Cubuk, Efthimios Kaxiras, Andrea J. Liu

Keywords: glass transition

Abstract: The dynamical glass transition is typically taken to be the temperature at which a glassy liquid is no longer able to equilibrate on experimental timescales. Consequently, the physical properties of these systems just above or below the dynamical glass transition, such as viscosity, can change by many orders of magnitude over long periods of time following external perturbation. During this progress toward equilibrium, glassy systems exhibit a history dependence that has complicated their study. In previous work, we bridged the gap between structure and dynamics in glassy liquids above their dynamical glass transition temperatures by introducing a scalar field called "softness," a quantity obtained using machine-learning methods. Softness is designed to capture the hidden patterns in relative particle positions that correlate strongly with dynamical rearrangements of particle positions. Here we show that the out-of-equilibrium behavior of a model glass-forming system can be understood

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors
Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article

Peer Reviewed: Y

Publication Status: 1-Published

Journal: Science

Publication Identifier Type: DOI

Publication Identifier: 10.1126/science.aal2538

Volume: 355

Issue: 6324

First Page #: 503

Date Submitted: 8/16/18 12:00AM

Date Published: 2/1/17 3:00PM

Publication Location:

Article Title: Magnetic resonance spectroscopy of an atomically thin material using a single-spin qubit

Authors: I. Lovchinsky, J. D. Sanchez-Yamagishi, E. K. Urbach, S. Choi, S. Fang, T. I. Andersen, K. Watanabe, T.

Keywords: Nitrogen Vacancy, qubit, boron nitride

Abstract: Two-dimensional (2D) materials offer a promising platform for exploring condensed matter phenomena and developing technological applications. However, the reduction of material dimensions to the atomic scale poses a challenge for traditional measurement and interfacing techniques that typically couple to macroscopic observables. We demonstrate a method for probing the properties of 2D materials via nanometer-scale nuclear quadrupole resonance (NQR) spectroscopy using individual atomlike impurities in diamond. Coherent manipulation of shallow nitrogen-vacancy (NV) color centers enables the probing of nanoscale ensembles down to approximately 30 nuclear spins in atomically thin hexagonal boron nitride (h-BN). The characterization of low-dimensional nanoscale materials could enable the development of new quantum hybrid systems, combining atomlike systems coherently coupled with individual atoms in 2D materials.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

Publication Type: Journal Article

Peer Reviewed: Y

Publication Status: 1-Published

Journal: Physical Review B

Publication Identifier Type: DOI

Publication Identifier: 10.1103/PhysRevB.95.014106

Volume: 95

Issue: 1

First Page #:

Date Submitted: 8/16/18 12:00AM

Date Published: 1/1/17 3:00PM

Publication Location:

Article Title: Strain effects on the behavior of isolated and paired sulfur vacancy defects in monolayer

Authors: Mehmet Gokhan Sensoy, Dmitry Vinichenko, Wei Chen, Cynthia M. Friend, Efthimios Kaxiras

Keywords: DFT, defects, MoS₂, TMDC

Abstract: We investigate the behavior of sulfur vacancy defects, the most abundant type of intrinsic defect in monolayer MoS₂, using first-principles calculations based on density functional theory. We consider the dependence of the isolated defect formation energy on the charge state and on uniaxial tensile and compressive strain up to 5%. We also consider the possibility of defect clustering by examining the formation energies of pairs of vacancies at various relative positions, and their dependence on charge state and strain. We find that there is no driving force for vacancy clustering, independent of strain in the material. The barrier for diffusion of S vacancies is larger than 1.9 eV in both charged and neutral states regardless of the presence of other nearby vacancies. We conclude that the formation of extended defects from S vacancies in planar monolayer MoS₂ is hindered both thermodynamically and kinetically.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: The Journal of Physical Chemistry C

Publication Identifier Type: DOI

Publication Identifier: 10.1021/acs.jpcc.6b11653

Volume: 121

Issue: 1

First Page #: 609

Date Submitted: 8/16/18 12:00AM

Date Published: 1/1/17 10:00AM

Publication Location:

Article Title: Polyiodide-Doped Graphene

Authors: Robert A. Hoyt, E. Marielle Remillard, Ekin D. Cubuk, Chad D. Vecitis, Efthimios Kaxiras

Keywords: DFT, dopant, graphene

Abstract: Iodine-doped graphene has recently attracted significant interest as a result of its enhanced conductivity and improved catalytic activity. Using density functional theory calculations, we obtain the formation energy, desorption rate, and electronic properties for graphene systems doped with polyiodide chains consisting of 1–6 iodine atoms in the low-concentration limit. We find that I3 and I5 act as p-type surface dopants that shift the Fermi level 0.46 and 0.57 eV below the Dirac point, respectively. For these two molecules, molecular orbital theory and analysis of the charge density show that doping transfers electronic charge to iodine π^* molecular orbitals oriented perpendicular to the graphene sheet. For even-length polyiodides, we find that I6 and I4 decompose to I2, which readily desorbs at 300 K. Adsorption energy calculations further show that I3 acts as an effective catalyst for the oxygen reduction reaction on graphene by stabilizing the rate-limiting OOH intermediate.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

Publication Type: Journal Article

Peer Reviewed: Y

Publication Status: 1-Published

Journal: 2D Materials

Publication Identifier Type: DOI

Publication Identifier: 10.1088/2053-1583/aa8313

Volume: 4

Issue: 4

First Page #: 045001

Date Submitted: 8/8/18 12:00AM

Date Published: 8/17/17 8:00AM

Publication Location:

Article Title: Properties of in-plane graphene/MoS2 heterojunctions

Authors: Wei Chen, Yuan Yang, Zhenyu Zhang, Efthimios Kaxiras

Keywords: Graphene, MoS2, heterojunction, DFT

Abstract: The graphene/MoS2 heterojunction formed by joining the two components laterally in a single plane promises to exhibit a low-resistance contact according to the Schottky–Mott rule. Here we provide an atomic-scale description of the structural, electronic, and magnetic properties of this type of junction. We first identify the energetically favorable structures in which the preference of forming C–S or C–Mo bonds at the boundary depends on the chemical conditions. We find that significant non-carrier related charge transfer between graphene and undoped MoS2 is localized at the boundary. We show that the abundant 1D boundary states substantially pin the Fermi level in the lateral contact between graphene and MoS2, in close analogy to the effect of 2D interfacial states in the contacts between 3D materials. Furthermore, we propose specific ways in which these effects can be exploited to achieve spin-polarized currents.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

RPPR Final Report
as of 16-Mar-2022

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Physical Review B

Publication Identifier Type: DOI

Publication Identifier: 10.1103/PhysRevB.96.245111

Volume: 96

Issue: 24

First Page #:

Date Submitted: 8/13/18 12:00AM

Date Published: 12/8/17 5:00AM

Publication Location:

Article Title: Effects of magnetic dopants in (Li_{0.8}M_{0.2}OH)FeSe (M = Fe, Mn, Co): Density functional theory study using a band unfolding technique

Authors: M. X. Chen, Wei Chen, Zhenyu Zhang, M. Weinert

Keywords: DFT, Surface States, Dopants

Abstract: The effects of Fe dopants on the electronic bands structure of (Li_{0.8}Fe_{0.2}OH)FeSe are investigated by a band unfolding (k-projection) technique and first-principles supercell calculations. Doping 20% Fe into the LiOH layers causes electron donation to the FeSe layers, significantly changing the profile of bands around the Fermi level. Because of the weak bonding between the LiOH and FeSe layers the magnetic configuration of the dopants has only minor effects on the band structure. The electronic bands for the surface FeSe layer of (Li_{0.8}Fe_{0.2}OH)FeSe show noticeable differences compared to those of the inner layers, both in the location of the Fermi level and in details of the bands near the high symmetry points, resulting from different effective doping levels and the broken symmetry at the surface. The band structure for the surface FeSe layer with checkerboard antiferromagnetic order is reasonably consistent with angle-resolved photoemission results...

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Physical Review B

Publication Identifier Type: DOI

Publication Identifier: 10.1103/PhysRevB.98.085406

Volume: 98

Issue: 8

First Page #:

Date Submitted: 8/8/18 12:00AM

Date Published: 8/3/18 8:00AM

Publication Location:

Article Title: Raman spectrum of CrI₃: An ab-initio study

Authors: Daniel T. Larson, Efthimios Kaxiras

Keywords: Magnetism, Raman, CrI₃, TMTC

Abstract: We study the Raman spectrum of CrI₃, a material that exhibits magnetism in a single layer. We employ first-principles calculations within density functional theory to determine the effects of polarization, strain, and incident angle on the phonon spectra of the three-dimensional bulk and the single-layer two-dimensional structure, for both the high- and low-temperature crystal structures. Our results are in good agreement with existing experimental measurements and serve as a guide for additional investigations to elucidate the physics of this interesting material.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article

Peer Reviewed: Y

Publication Status: 1-Published

Journal: Nature

Publication Identifier Type: DOI

Publication Identifier: 10.1038/s41586-018-0205-0

Volume: 558

Issue: 7710

First Page #: 425

Date Submitted: 8/8/18 12:00AM

Date Published: 6/20/18 8:00AM

Publication Location:

Article Title: Heterointerface effects in the electrointercalation of van der Waals heterostructures

Authors: D. Kwabena Bediako, Mehdi Rezaee, Hyobin Yoo, Daniel T. Larson, S. Y. Frank Zhao, Takashi Taniguchi

Keywords: Intercalation, TMDC, graphene

Abstract: Molecular-scale manipulation of electronic and ionic charge accumulation in materials is the backbone of electrochemical energy storage. Layered van der Waals (vdW) crystals are a diverse family of materials into which mobile ions can electrochemically intercalate into the interlamellar gaps of the host atomic lattice^{5,6}. The structural diversity of such materials enables the interfacial properties of composites to be optimized to improve ion intercalation for energy storage and electronic devices. However, the ability of heterolayers to modify intercalation reactions, and their role at the atomic level, are yet to be elucidated. Here we demonstrate the electrointercalation of lithium at the level of individual atomic interfaces of dissimilar vdW layers. Electrochemical devices based on vdW heterostructures¹³ of stacked hexagonal boron nitride, graphene and molybdenum dichalcogenide (MoX₂; X=S, Se) layers are constructed ...

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

Publication Type: Journal Article

Peer Reviewed: Y

Publication Status: 1-Published

Journal: Nature

Publication Identifier Type: DOI

Publication Identifier: 10.1038/nature26160

Volume: 556

Issue: 7699

First Page #: 43

Date Submitted: 8/8/18 12:00AM

Date Published: 4/5/18 4:00AM

Publication Location:

Article Title: Unconventional superconductivity in magic-angle graphene superlattices

Authors: Yuan Cao, Valla Fatemi, Shiang Fang, Kenji Watanabe, Takashi Taniguchi, Efthymios Kaxiras, Pablo Ja

Keywords: Graphene, Superconductivity

Abstract: The behaviour of strongly correlated materials, and in particular unconventional superconductors, has been studied extensively for decades, but is still not well understood. This lack of theoretical understanding has motivated the development of experimental techniques for studying such behaviour, such as using ultracold atom lattices to simulate quantum materials. Here we report the realization of intrinsic unconventional superconductivity—which cannot be explained by weak electron–phonon interactions—in a two-dimensional superlattice created by stacking two sheets of graphene that are twisted relative to each other by a small angle. For twist angles of about 1.1°—the first ‘magic’ angle—the electronic band structure of this ‘twisted bilayer graphene’ exhibits flat bands near zero Fermi energy, resulting in correlated insulating states at half-filling. Upon electrostatic doping of the material away from these correlated insulating states, we observe tunable zero-resistance states ...

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article

Peer Reviewed: Y

Publication Status: 1-Published

Journal: Nature

Publication Identifier Type: DOI

Publication Identifier: 10.1038/nature26154

Volume: 556

Issue: 7699

First Page #: 80

Date Submitted: 8/8/18 12:00AM

Date Published: 4/5/18 4:00AM

Publication Location:

Article Title: Correlated insulator behaviour at half-filling in magic-angle graphene superlattices

Authors: Yuan Cao, Valla Fatemi, Ahmet Demir, Shiang Fang, Spencer L. Tomarken, Jason Y. Luo, Javier D. San

Keywords: Graphene, Superconductivity

Abstract: The behaviour of strongly correlated materials, and in particular unconventional superconductors, has been studied extensively for decades, but is still not well understood. This lack of theoretical understanding has motivated the development of experimental techniques for studying such behaviour, such as using ultracold atom lattices to simulate quantum materials. Here we report the realization of intrinsic unconventional superconductivity—which cannot be explained by weak electron–phonon interactions—in a two-dimensional superlattice created by stacking two sheets of graphene that are twisted relative to each other by a small angle. For twist angles of about 1.1° —the first ‘magic’ angle—the electronic band structure of this ‘twisted bilayer graphene’ exhibits flat bands near zero Fermi energy, resulting in correlated insulating states at half-filling. Upon electrostatic doping of the material away from these correlated insulating states, we observe tunable zero-resistance states ...

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

Publication Type: Journal Article

Peer Reviewed: Y

Publication Status: 1-Published

Journal: Physical Review B

Publication Identifier Type: Other

Publication Identifier:

Volume:

Issue:

First Page #:

Date Submitted: 8/18/18 12:00AM

Date Published:

Publication Location:

Article Title: Pressure dependence of the magic twist angle in graphene superlattices

Authors: Stephen Carr, Shiang Fang, Pablo Jarillo-Herrero, Efthimios Kaxiras

Keywords: Graphene, twistrionics, superconductivity, DFT

Abstract: The recently demonstrated unconventional superconductivity in twisted bilayer graphene (tBLG) opens the possibility for interesting applications of two-dimensional layers that involve correlated electron states. Here we explore the possibility of modifying electronic correlations by the application of uniaxial pressure on the weakly interacting layers, which results in increased interlayer coupling and a modification of the magic angle value and associated density of states. Our findings are based on first-principles calculations that accurately describe the height-dependent interlayer coupling through the combined use of Density Functional Theory and Maximally localized Wannier functions. We obtain the relationship between twist angle and external pressure for the magic angle flat bands of tBLG. This may provide a convenient method to tune electron correlations by controlling the length scale of the superlattice.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: Physical Review B
Publication Identifier Type: DOI **Publication Identifier:** 10.1103/PhysRevB.98.075106
Volume: 98 **Issue:** 7 **First Page #:**
Date Submitted: 8/8/18 12:00AM **Date Published:** 8/6/18 4:00AM
Publication Location:

Article Title: Electronic structure theory of strained two-dimensional materials with hexagonal symmetry

Authors: Shiang Fang, Stephen Carr, Miguel A. Cazalilla, Efthimios Kaxiras

Keywords: DFT, Graphene, TDMC, Strain

Abstract: We derive electronic tight-binding Hamiltonians for strained graphene, hexagonal boron nitride, and transition-metal dichalcogenides based on Wannier transformation of ab initio density functional theory calculations. Our microscopic models include strain effects to leading order that respect the hexagonal crystal symmetry and local crystal configuration and are beyond the central force approximation which assumes only pairwise distance dependence. Based on these models, we also derive and analyze the effective low-energy Hamiltonians. Our ab initio approaches complement the symmetry group representation construction for such effective low-energy Hamiltonians and provide the values of the coefficients for each symmetry-allowed term. These models are relevant for the design of electronic device applications since they provide the framework for describing the coupling of electrons to other degrees of freedom including phonons, spin, and the electromagnetic field. The models can also serv

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors
Acknowledged Federal Support: Y

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: Physical Review B
Publication Identifier Type: DOI **Publication Identifier:** 10.1103/PhysRevB.98.035203
Volume: 98 **Issue:** 3 **First Page #:**
Date Submitted: 8/8/18 12:00AM **Date Published:** 7/20/18 4:00AM
Publication Location:

Article Title: Enhanced superconductivity upon weakening of charge density wave transport in 2H-TaS₂ in the two-dimensional limit

Authors: Yafang Yang, Shiang Fang, Valla Fatemi, Jonathan Ruhman, Efrén Navarro-Moratalla, Kenji Watanabe,

Keywords: TMDC, Superconductivity, DFT, Charge density waves

Abstract: Layered transition-metal dichalcogenides that host coexisting charge-density wave (CDW) and superconducting orders provide ideal systems for exploring the effects of dimensionality on correlated electronic phases. Dimensionality has a profound effect on both superconductivity and CDW instabilities. Here we report a substantial enhancement of the superconducting T_c to 3.4 K for 2H-TaS₂ in the monolayer limit, compared to 0.8 K in the bulk. In addition, the transport signature of a CDW phase transition vanishes in the two-dimensional limit. In our analysis of electronic and vibrational properties of this material, we show that a reduction of the CDW amplitude results in a substantial increase of the density of states at the Fermi energy, which can boost T_c by an amount similar to that seen in experiment. Our results indicate competition between CDW order and superconductivity in ultrathin 2H-TaS₂ down to the monolayer limit, providing insight toward understanding ...

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors
Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 4-Under Review
Journal: Physical Review B
Publication Identifier Type: Other **Publication Identifier:**
Volume: **Issue:** **First Page #:**
Date Submitted: 8/13/18 12:00AM **Date Published:**
Publication Location:
Article Title: The Berry curvature dipole current in transition metal dichalcogenides family
Authors: Jhih-Shih You, Shiang Fang, Su-Yang Xu, Efthimios Kaxiras, Tony Low
Keywords: DFT, Berry curvature, TMDC
Abstract: We study the quantum nonlinear Hall effect in two-dimensional materials with time-reversal symmetry. When only one mirror line exists, a transverse charge current occurs in second-order response to an external electric field, as a result of the Berry curvature dipole in momentum space. Candidate 2D materials to observe this effect are two-dimensional transition-metal dichalcogenides~(TMDCs). First we use an ab initio based tight-binding approach to demonstrate that monolayer Td-structure TMDCs exhibit a finite Berry curvature dipole. In the 1H and 1T? phase of TMDCs, we show the emergence of finite Berry curvature dipole with the application of strain and electrical displacement field respectively.
Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors
Acknowledged Federal Support: Y

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: Computer Physics Communications
Publication Identifier Type: DOI **Publication Identifier:** 10.1016/j.cpc.2018.04.011
Volume: 230 **Issue:** **First Page #:** 27
Date Submitted: 8/8/18 12:00AM **Date Published:** 9/1/18 4:00AM
Publication Location:
Article Title: Compression of Wannier functions into Gaussian-type orbitals
Authors: Athmane Bakhta, Eric Cancès, Paul Cazeaux, Shiang Fang, Efthimios Kaxiras
Keywords: Wannier functions, Gaussian-type orbitals, 2D materials, Greedy algorithms
Abstract: We propose a greedy algorithm for the compression of Wannier functions into Gaussian-polynomials orbitals. The so-obtained compressed Wannier functions can be stored in a very compact form, and can be used to efficiently parameterize effective tight-binding Hamiltonians for multilayer 2D materials for instance. The compression method preserves the symmetries (if any) of the original Wannier function. We provide algorithmic details, and illustrate the performance of our implementation on several examples, including graphene, hexagonal boron-nitride, single-layer FeSe, and bulk silicon in the diamond cubic structure.
Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors
Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: NONE

Publication Identifier Type:

Publication Identifier:

Volume:

Issue:

First Page #:

Date Submitted: 8/14/19 12:00AM

Date Published:

Publication Location:

Article Title: Data-driven studies of magnetic two-dimensional materials

Authors: Trevor David Rhone, Wei Chen, Shaan Desai, Amir Yacoby, Efthimios Kaxiras

Keywords: Machine Learning, TMTC, DFT

Abstract: We use a data-driven approach to study the magnetic and thermodynamic properties of van der Waals (vdW) layered materials. We investigate monolayers of the form $A_2B_2X_6$, based on the known material $Cr_2Ge_2Te_6$, using density functional theory (DFT) calculations and machine learning methods to determine their magnetic properties, such as magnetic order and magnetic moment. We also examine formation energies and use them as a proxy for chemical stability. We show that machine learning tools, combined with DFT calculations, can provide a computationally efficient means to predict properties of such two-dimensional (2D) magnetic materials. Our data analytics approach provides insights into the microscopic origins of magnetic ordering in these systems. For instance, we find that the X site strongly affects the magnetic coupling between neighboring A sites, which drives the magnetic ordering. Our approach opens new ways for rapid discovery of chemically stable vdW materials that

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Physical Review B

Publication Identifier Type:

Publication Identifier: 10.1103/PhysRevB.98.224102

Volume: 98

Issue:

First Page #: 224102

Date Submitted: 7/10/19 12:00AM

Date Published: 12/5/18 5:00AM

Publication Location:

Article Title: Relaxation and Domain Formation in Incommensurate 2D Heterostructures

Authors: Stephen Carr, Daniel Massatt, Steven B. Torrisi, Paul Cazeaux, Mitchell Luskin, Efthimios Kaxiras

Keywords: Relaxation, Bilayer, Graphene, TMDC

Abstract: We introduce configuration space as a natural representation for calculating the mechanical relaxation patterns of incommensurate two-dimensional (2D) bilayers, bypassing supercell approximations to encompass aperiodic relaxation patterns. The approach can be applied to a wide variety of 2D materials through the use of a continuum model in combination with a generalized stacking fault energy for interlayer interactions. We present computational results for small-angle twisted bilayer graphene and molybdenum disulfide (MoS_2), a representative material of the transition metal dichalcogenide (TMDC) family of 2D semiconductors. We calculate accurate relaxations for MoS_2 even at small twist-angle values, enabled by the fact that our approach does not rely on empirical atomistic potentials for interlayer coupling. The results demonstrate the efficiency of the configuration space method by computing relaxations with minimal computational cost for twist angles down to 0.05° ...

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: Multiscale Modeling & Simulation
Publication Identifier Type: DOI **Publication Identifier:** 10.1137/17M1141035
Volume: 16 **Issue:** 1 **First Page #:** 429
Date Submitted: 8/8/18 12:00AM **Date Published:** 3/6/18 5:00AM
Publication Location:

Article Title: Incommensurate Heterostructures in Momentum Space

Authors: Daniel Massatt, Stephen Carr, Mitchell Luskin, Christoph Ortner

Keywords: momentum space, two-dimensional materials, electronic structure, density of states, heterostructure

Abstract: To make the investigation of electronic structure of incommensurate heterostructures computationally tractable, effective alternatives to Bloch theory must be developed. In [Multiscale Model. Simul., 15(2017), pp. 476--499] we developed and analyzed a real space scheme that exploits spatial ergodicity and near-sightedness. In the present work, we present an analogous scheme formulated in momentum space, which we prove has significant computational advantages in specific incommensurate systems of physical interest, e.g., bilayers of a specified class of materials with small rotation angles. We use our theoretical analysis to obtain estimates for improved rates of convergence with respect to total CPU time for our momentum space method that are confirmed in computational experiments.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 4-Under Review
Journal: NONE
Publication Identifier Type: **Publication Identifier:**
Volume: **Issue:** **First Page #:**
Date Submitted: 8/13/18 12:00AM **Date Published:**
Publication Location:

Article Title: Density functional theory beyond the Born-Oppenheimer approximation: exact treatment of the ionic zero-point motion

Authors: Grigory Kolesov, Efthimios Kaxiras, Efstratios Manousakis

Keywords: DFT, Path integral, Born-Oppenheimer

Abstract: We introduce a method to carry out zero-temperature calculations within density functional theory (DFT) but without relying on the Born-Oppenheimer (BO) approximation for the ionic motion. Our approach is based on the finite-temperature many-body path-integral formulation of quantum mechanics by taking the zero-temperature limit and treating the imaginary-time propagation of the electronic variables exactly in the context of DFT. This goes beyond the familiar BO approximation and is limited from being an exact treatment of both electrons and ions only by the approximations involved in the DFT component. We test our method in two simple molecules, H₂ and benzene. We demonstrate that the method produces a difference from the results of the BO approximation which is significant for many physical systems, especially those containing light atoms such as hydrogen; in these cases, we find that the fluctuations of the distance from its equilibrium position, due to the zero-point-motion ...

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: Nano Letters
Publication Identifier Type: DOI **Publication Identifier:** 10.1021/acs.nanolett.7b02500
Volume: 17 **Issue:** 10 **First Page #:** 6077
Date Submitted: 8/8/18 12:00AM **Date Published:** 9/5/17 4:00AM
Publication Location:

Article Title: Reassessing Graphene Absorption and Emission Spectroscopy

Authors: Yuan Yang, Grigory Kolesov, Lucas Kocia, Eric J. Heller

Keywords: condensed matter spectroscopy, Graphene, optical absorption, optical emission, UV-vis spectroscopy

Abstract: We present a new paradigm for understanding optical absorption and hot electron dynamics experiments in graphene. Our analysis pivots on assigning proper importance to phonon-assisted indirect processes and bleaching of direct processes. We show indirect processes figure in the excess absorption in the UV region. Experiments which were thought to indicate ultrafast relaxation of electrons and holes, reaching a thermal distribution from an extremely nonthermal one in under 5–10 fs, instead are explained by the nascent electron and hole distributions produced by indirect transitions. These need no relaxation or ad-hoc energy removal to agree with the observed emission spectra and fast pulsed absorption spectra. The fast emission following pulsed absorption is dominated by phonon-assisted processes, which vastly outnumber direct ones and are always available, connecting any electron with any hole any time. Calculations are given, including explicitly calculating the magnitude of indirect

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 0-Other
Journal: Studies in Applied Mathematics
Publication Identifier Type: DOI **Publication Identifier:** 10.1111/sapm.12180
Volume: 139 **Issue:** 4 **First Page #:** 599
Date Submitted: 8/9/18 12:00AM **Date Published:** 11/1/17 5:00AM
Publication Location:

Article Title: On the Wiener-Hopf Method for Surface Plasmons: Diffraction from Semiinfinite Metamaterial Sheet

Authors: Dionisios Margetis, Matthias Maier, Mitchell Luskin

Keywords: Wiener-Hopf

Abstract: By formally invoking the Wiener–Hopf method, we explicitly solve a one-dimensional, singular integral equation for the excitation of a slowly decaying electromagnetic wave, called surface plasmon-polariton (SPP), of small wavelength on a semiinfinite, flat conducting sheet irradiated by a plane wave in two spatial dimensions. This setting is germane to wave diffraction by edges of large sheets of single-layer graphene. Our analytical approach includes (i) formulation of a functional equation in the Fourier domain; (ii) evaluation of a split function, which is expressed by a contour integral and is a key ingredient of the Wiener–Hopf factorization; and (iii) extraction of the SPP as a simple-pole residue of a Fourier integral. Our analytical solution is in good agreement with a finite-element numerical computation.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: ACS Photonics

Publication Identifier Type: DOI

Publication Identifier: 10.1021/acsp Photonics.7b01094

Volume: 5

Issue: 2

First Page #: 544

Date Submitted: 8/9/18 12:00AM

Date Published: 12/1/17 6:00AM

Publication Location:

Article Title: Ultracompact Amplitude Modulator by Coupling Hyperbolic Polaritons over a Graphene-Covered Gap

Authors: Matthias Maier, Andrei Nemilentsau, Tony Low, Mitchell Lusk

Keywords: hyperbolic polariton

Abstract: The hyperbolic phonon-polaritons within the Reststrahlen band of hBN are of great interest for applications in nanophotonics, as they are capable of propagating light signals with low losses over large distances. However, due to the phononic nature of the polaritons in hBN, amplitude modulation of its signal proves to be difficult and has been underexplored. In this paper, we propose theoretically a broadband efficient amplitude modulator for hyperbolic rays in hBN operating in the frequency range between 1450 and 1550 cm⁻¹. The modulating region comprises a few tens of nanometers wide gap carved within the hBN slab and covered by a graphene layer, where electrostatically gated graphene serves as a mediator that facilitates the coupling between phonon-polaritons on each side of the gap through plasmonic modes within graphene. We demonstrate that such an ultracompact modulator has insertion losses as low as 3 dB and provides modulation depth varying between 14 and 20 dB within the type-I

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Physical Review B

Publication Identifier Type: DOI

Publication Identifier: 10.1103/PhysRevB.97.035307

Volume: 97

Issue: 3

First Page #:

Date Submitted: 8/9/18 12:00AM

Date Published: 1/1/18 12:00AM

Publication Location:

Article Title: Universal behavior of a dispersive Dirac cone in gradient-index plasmonic metamaterials

Authors: Matthias Maier, Marios Mattheakis, Efthimios Kaxiras, Mitchell Lusk, Dionisios Margetis

Keywords: metamaterial

Abstract: We demonstrate analytically and numerically that the dispersive Dirac cone emulating an epsilon-near-zero (ENZ) behavior is a universal property within a family of plasmonic crystals consisting of two-dimensional (2D) metals. Our starting point is a periodic array of 2D metallic sheets embedded in an inhomogeneous and anisotropic dielectric host that allows for propagation of transverse-magnetic (TM) polarized waves. By invoking a systematic bifurcation argument for arbitrary dielectric profiles in one spatial dimension, we show how TM Bloch waves experience an effective dielectric function that averages out microscopic details of the host medium. The corresponding effective dispersion relation reduces to a Dirac cone when the conductivity of the metallic sheet and the period of the array satisfy a critical condition for ENZ behavior. Our analytical findings are in excellent agreement with numerical simulations.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Communications in Mathematical Sciences

Publication Identifier Type: DOI

Publication Identifier: 10.4310/CMS.2018.v16.n1.a4

Volume: 16

Issue: 1

First Page #: 77

Date Submitted: 8/9/18 12:00AM

Date Published:

Publication Location:

Article Title: Generation of surface plasmon-polaritons by edge effects

Authors: Matthias Maier, Dionisios Margetis, Mitchell Luskin

Keywords: time-harmonic Maxwell's equations, finite element method, surface plasmon-polariton, singular geometry, weak discontinuity on hypersurface.

Abstract: By using numerical and analytical methods, we describe the generation of fine-scale lateral electromagnetic waves, called surface plasmon-polaritons (SPPs), on atomically thick, metamaterial conducting sheets in two spatial dimensions (2D). Our computations capture the two-scale character of the total field and reveal how each edge of the sheet acts as a source of an SPP that may dominate the diffracted field. We use the finite element method to numerically implement a variational formulation for a weak discontinuity of the tangential magnetic field across a hypersurface. An adaptive, local mesh refinement strategy based on a posteriori error estimators is applied to resolve the pronounced two-scale character of wave propagation and radiation over the metamaterial sheet. We demonstrate by numerical examples how a singular geometry, e.g., sheets with sharp edges, and sharp spatial changes in the associated surface conductivity may significantly influence surface plasmons in nanophotonics.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

Publication Type: Journal Article

Peer Reviewed: Y

Publication Status: 1-Published

Journal: Journal of Applied Physics

Publication Identifier Type: DOI

Publication Identifier: 10.1063/1.5007842

Volume: 122

Issue: 24

First Page #: 244301

Date Submitted: 8/14/18 12:00AM

Date Published: 12/1/17 6:00AM

Publication Location:

Article Title: A force-matching Stillinger-Weber potential for MoS

Authors: Mingjian Wen, Sharmila N. Shirodkar, Petr Plecháč, Efthimios Kaxiras, Ryan S. Elliott, Eyal B. Tadmor

Keywords: interatomic potentials, molybdenum disulphide

Abstract: Two-dimensional molybdenum disulfide (MoS₂) is a promising material for the next generation of switchable transistors and photodetectors. In order to perform large-scale molecular simulations of the mechanical and thermal behavior of MoS₂-based devices, an accurate interatomic potential is required. To this end, we have developed a Stillinger-Weber potential for monolayer MoS₂. The potential parameters are optimized to reproduce the geometry (bond lengths and bond angles) of MoS₂ in its equilibrium state and to match as closely as possible the forces acting on the atoms along a dynamical trajectory obtained from ab initio molecular dynamics. Verification calculations indicate that the new potential accurately predicts important material properties including the strain dependence of the cohesive energy, the elastic constants, and the linear thermal expansion coefficient. The uncertainty in the potential parameters is determined using a Fisher information theory analysis. It is found that

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Journal of the Mechanics and Physics of Solids

Publication Identifier Type: DOI

Publication Identifier: 10.1016/j.jmps.2017.12.005

Volume: 112

Issue:

First Page #: 225

Date Submitted: 8/14/18 12:00AM

Date Published: 8/14/18 10:00AM

Publication Location:

Article Title: Structural and electron diffraction scaling of twisted graphene bilayers

Authors: Kuan Zhang, Ellad B. Tadmor

Keywords: Twisted graphene bilayers, moire pattern, electron diffraction, multiscale simulation

Abstract: Multiscale simulations are used to study the structural relaxation in twisted graphene bi- layers and the associated electron diffraction patterns. The initial twist forms an incommensurate moire pattern that relaxes to a commensurate microstructure comprised of a repeating pattern of alternating low-energy AB and BA domains surrounding a high-energy AA domain. The simulations show that the relaxation mechanism involves a localized rotation and shrinking of the AA domains that scales in two regimes with the imposed twist. For small twisting angles, the localized rotation tends to a constant; for large twist, the rotation scales linearly with it. This behavior is tied to the inverse scaling of the moire pattern size with twist angle and is explained theoretically using a linear elasticity model. The results are validated experimentally through a simulated electron diffraction analysis of the relaxed structures. A complex electron diffraction pattern involving the appearance of weak

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

Publication Type: Journal Article

Peer Reviewed: Y

Publication Status: 1-Published

Journal: Ann. Henri Poincaré

Publication Identifier Type: DOI

Publication Identifier: <https://doi.org/10.1007/s00023-018-0699-x>

Volume:

Issue:

First Page #: 1

Date Submitted: 8/17/18 12:00AM

Date Published: 7/2/18 4:00AM

Publication Location:

Article Title: Canonical Quantum Observables for Molecular Systems Approximated by Ab Initio Molecular Dynamics

Authors: A. Kammonen, P. Plechac, M. Sandberg, A. Szepessy

Keywords: Born-Oppenheimer approximation, canonical ensemble, Weyl quantization, ab initio molecular dynamics, quantum observables

Abstract: It is known that ab initio molecular dynamics based on the electron ground-state eigenvalue can be used to approximate quantum observables in the canonical ensemble when the temperature is low compared to the first electron eigenvalue gap. This work proves that a certain weighted average of the different ab initio dynamics, corresponding to each electron eigenvalue, approximates quantum observables for any temperature. The proof uses the semiclassical Weyl law to show that canonical quantum observables of nuclei-electron systems, based on matrix-valued Hamiltonian symbols, can be approximated by ab initio molecular dynamics with the error proportional to the electron-nuclei mass ratio. The result covers observables that depend on time correlations. A combination of the Hilbert-Schmidt inner product for quantum operators and Weyl's law shows that the error estimate holds for observables and Hamiltonian symbols that have three and five bounded derivatives, respectively, provided

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Journal of Physics: Materials

Publication Identifier Type: DOI

Publication Identifier:

Volume:

Issue:

First Page #:

Date Submitted: 8/13/19 12:00AM

Date Published:

Publication Location:

Article Title: Deciphering the origin of nonlocal resistance in multiterminal graphene on hexagonal-boron-nitride with ab initio quantum transport: Fermi surface edge currents rather than Fermi sea topological valley currents

Authors: M. Marmolejo-Tejada, J. H. Garc a, M. Petrovic, P.-H. Chang, X.-L. Sheng, A. Cresti, P. Plechac, S. Ro

Keywords: graphene on hBN, non-local resistance, quantum transport

Abstract: The recent observation (Gorbachev et al 2014 Science 346 448) of nonlocal resistance R near the Dirac point (DP) of multiterminal graphene on aligned hexagonal-boron nitride (G/hBN) has been interpreted as the consequence of topological valley Hall currents carried by the Fermi sea states just beneath the bulk gap E_g induced by the inversion symmetry breaking. However, the corresponding valley Hall conductivity, quantized inside E_g , is not directly measurable. Conversely, the Landauer–B ttiker formula, as numerically exact approach to observable nonlocal transport quantities, yields $R \neq 0$ for the same simplistic Hamiltonian of the gapped graphene that generates non-zero valley Hall conductivity via the Kubo formula. We combine ab initio with quantum transport calculations to demonstrate that G/hBN wires with zigzag edges host dispersive edge states near the DP that are absent in theories based on the simplistic Hamiltonian.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Nano Letters

Publication Identifier Type:

Publication Identifier:

Volume:

Issue:

First Page #:

Date Submitted: 8/18/18 12:00AM

Date Published:

Publication Location:

Article Title: Spatio-temporal dynamics of shift current quantum pumping by femtosecond light pulse,

Authors: U. Bajpai, B. S. Popescu, P. Plechac, B.K. Nikolic, L. E. F. Foa Torres, H. Ishizuka, N. Nagaosa

Keywords: Nonlinear optics, ultrafast phenomena, photovoltaics, optoelectronics, nonadiabatic quantum pumping in nanostructures, time-dependent quantum transport simulations

Abstract: We analyze realistic two-terminal devices, where paradigmatic Rice-Mele model is sandwiched between two metallic electrodes, using recently developed time-dependent nonequilibrium Green function algorithms scaling linearly in the number of time steps and capable of treating nonperturbative effects in the amplitude of external time-dependent fields. This unveils novel features: superballistic transport, signified by time dependence of the displacement, $\langle x \rangle \propto t^\alpha$ with $\alpha > 1$, of the photoexcited charge carriers from the region where the femtosecond light pulse is applied toward the electrodes; and photocurrent quadratic in light intensity at subgap frequencies of light due to two-photon absorption processes that were missed in previous perturbative analyses. Furthermore, frequency dependence of the DC component of the photocurrent reveals shift currents as a realization of nonadiabatic quantum charge pumping enabled by breaking of left-right symmetry of the device structure.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: Physical Review Applied
Publication Identifier Type: **Publication Identifier:**
Volume: **Issue:** **First Page #:**
Date Submitted: 8/18/18 12:00AM **Date Published:**
Publication Location:

Article Title: Spin and charge pumping by steady or pulse current-driven magnetic domain wall: A self-consistent multiscale time-dependent-quantum/time-dependent-classical approach

Authors: M. Petrovic, B. S. Popescu, U. Bajpai, P. Plechac, B. K. Nikolic

Keywords: time-dependent non-equilibrium Green function, magnetic domain walls, Landau-Lifshitz-Gilbert equation, quantum transport

Abstract: We introduce a multiscale framework which combines time-dependent nonequilibrium Green function (TD-NEGF) algorithms, scaling linearly in the number of time steps and describing quantum-mechanically conduction electrons in the presence of time-dependent fields of arbitrary strength or frequency, with classical time evolution of localized magnetic moments described by the Landau-Lifshitz-Gilbert (LLG) equation. The TD-NEGF+LLG framework can be applied to a variety of problems where current-driven spin torque induces dynamics of magnetic moments as the key resource for next generation spintronics. Using magnetic domain wall (DW) as an example, we predict that its motion will pump time-dependent spin and charge currents (on the top of unpolarized DC charge current injected through normal metal leads to drive the DW motion). We also quantify the DW transient inertial displacement due to its acceleration and deceleration by pulse current and the entailed spin and charge pumping.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors
Acknowledged Federal Support: Y

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: Nano Letters
Publication Identifier Type: DOI **Publication Identifier:** 10.1021/acs.nanolett.7b04396
Volume: 18 **Issue:** 1 **First Page #:** 460
Date Submitted: 8/18/18 12:00AM **Date Published:** 12/1/17 12:00AM
Publication Location:

Article Title: Controlled Electrochemical Intercalation of Graphene/

Authors: S. Y. Frank Zhao, Giselle A. Elbaz, D. Kwabena Bediako, Cyndia Yu, Dmitri K. Efetov, Yinsheng Guo, J.

Keywords: Nanoscale electrochemistry, graphite intercalation, graphene, van der Waals heterostructures, host-guest

Abstract: Electrochemical intercalation is a powerful method for tuning the electronic properties of layered solids. In this work, we report an electrochemical strategy to controllably intercalate lithium ions into a series of van der Waals (vdW) heterostructures built by sandwiching graphene between hexagonal boron nitride (h-BN). We demonstrate that encapsulating graphene with h-BN eliminates parasitic surface side reactions while simultaneously creating a new heterointerface that permits intercalation between the atomically thin layers. To monitor the electrochemical process, we employ the Hall effect to precisely monitor the intercalation reaction. We also simultaneously probe the spectroscopic and electrical transport properties of the resulting intercalation compounds at different stages of intercalation. We achieve the highest carrier density $>5 \times 10^{13} \text{ cm}^{-2}$ with mobility $>103 \text{ cm}^2/(\text{V s})$ in the most heavily intercalated samples, where Shubnikov-de Haas quantum oscillations are observed at low

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors
Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article

Peer Reviewed: Y

Publication Status: 1-Published

Journal: Nature Nanotechnology

Publication Identifier Type: DOI

Publication Identifier: 10.1038/s41565-017-0030-x

Volume: 13

Issue: 2

First Page #: 128

Date Submitted: 8/18/18 12:00AM

Date Published: 1/1/18 12:00AM

Publication Location:

Article Title: Electrical control of charged carriers and excitons in atomically thin materials

Authors: Ke Wang, Kristiaan De Greve, Luis A. Jauregui, Andrey Sushko, Alexander High, You Zhou, Giovanni S

Keywords: charged carriers, excitons

Abstract: Electrical confinement and manipulation of charge carriers in semiconducting nanostructures are essential for realizing functional quantum electronic devices^{1–3}. The unique band structure^{4–7} of atomically thin transition metal dichalcogenides (TMDs) offers a new route towards realizing novel 2D quantum electronic devices, such as valleytronic devices and valley–spin qubits⁸. 2D TMDs also provide a platform for novel quantum optoelectronic devices^{9–11} due to their large exciton binding energy^{12,13}. However, controlled confinement and manipulation of electronic and excitonic excitations in TMD nanostructures have been technically challenging due to the prevailing disorder in the material, preventing accurate experimental control of local confinement and tunnel couplings^{14–16}. Here we demonstrate a novel method for creating high-quality heterostructures composed of atomically thin materials that allows for efficient electrical control of excitations. Specifically, we demonstrate quantum

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

Publication Type: Journal Article

Peer Reviewed: Y

Publication Status: 1-Published

Journal: Nanotechnology

Publication Identifier Type: DOI

Publication Identifier: 10.1088/1361-6528/aad79f

Volume: 29

Issue: 42

First Page #:

Date Submitted: 8/18/18 12:00AM

Date Published: 10/1/18 12:00AM

Publication Location:

Article Title: Imaging quantum dot formation in MoS

Authors: S Bhandari, K Wang, K Watanabe, T Taniguchi, P Kim, R M Westervelt

Keywords: scanning probe microscope, MoS₂, quantum dots, imaging quantum dots

Abstract: Among two-dimensional materials, semiconducting ultrathin sheets of MoS₂ are promising for nanoelectronics. We show how a scanning probe microscope (SPM) can be used to image the flow of electrons in a MoS₂ Hall bar sample at 4.2 K allowing us to understand device physics at the nanoscale. The SPM tip acts as a movable gate and capacitively couples the SPM tip to the device below. By measuring the change in device conductance as the tip is raster scanned across the sample, spatial maps of the device conductance can be obtained. We present images showing the characteristic ‘bullseye’ pattern of Coulomb blockade conductance rings around a quantum dot formed in a narrow contact as the carrier density is depleted with a backgate. These images show that multiple dots are created by the disorder potential in MoS₂. From these SPM images, we estimate the size and position of these quantum dots using a capacitive model.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Physical Review B

Publication Identifier Type: DOI

Publication Identifier: 10.1103/PhysRevB.98.195407

Volume: 98

Issue: 19

First Page #:

Date Submitted: 7/9/19 12:00AM

Date Published: 11/1/18 12:00AM

Publication Location:

Article Title: Nonperturbative nonlinear effects in the dispersion relations for TE and TM plasmons on two-dimensional materials

Authors: Vera Andreeva, Mitchell Luskin, Dionisios Margetis

Keywords: Waveguide configurations; Time-harmonic Maxwell's equations; Adaptive finite-element methods; Surface plasmon–polariton

Abstract: We discuss analytically and numerically the propagation and energy transmission of electromagnetic waves caused by the coupling of surface plasmon polaritons (SPPs) between two spatially separated layers of 2D materials, such as graphene, at subwavelength distances. We construct an adaptive finite-element method to compute the ratio of energy transmitted within these waveguide structures reliably and efficiently. At its heart, the method is built upon a goal-oriented a posteriori error estimation with the dual-weighted residual method (DWR). Furthermore, we derive analytic solutions of the two-layer system, compare those to (known) single-layer configurations, and compare and validate our numerical findings by comparing numerical and analytical values for optimal spacing of the two-layer configuration. Additional aspects of our numerical treatment, such as local grid refinement, and the utilization of perfectly matched layers (PMLs) are examined in detail.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Nature Materials

Publication Identifier Type: DOI

Publication Identifier: 10.1038/s41563-019-0346-z

Volume: 18

Issue: 5

First Page #: 448

Date Submitted: 7/9/19 12:00AM

Date Published: 4/1/19 5:00AM

Publication Location:

Article Title: Atomic and electronic reconstruction at the van der Waals interface in twisted bilayer graphene

Authors: Hyobin Yoo, Rebecca Engelke, Stephen Carr, Shiang Fang, Kuan Zhang, Paul Cazeaux, Suk Hyun Sur

Keywords: van der Waals interface, twisted bilayer graphene

Abstract: Control of the interlayer twist angle in two-dimensional van der Waals (vdW) heterostructures enables one to engineer a quasiperiodic moiré superlattice of tunable length scale^{1–8}. In twisted bilayer graphene, the simple moiré superlattice band description suggests that the electronic bandwidth can be tuned to be comparable to the vdW interlayer interaction at a 'magic angle'⁹, exhibiting strongly correlated behaviour. However, the vdW interlayer interaction can also cause significant structural reconstruction at the interface by favouring interlayer commensurability, which competes with the intralayer lattice distortion^{10–16}. Here we report atomicscale reconstruction in twisted bilayer graphene and its effect on the electronic structure.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

RPPR Final Report
as of 16-Mar-2022

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Computer Methods in Applied Mechanics and Engineering

Publication Identifier Type: DOI

Publication Identifier: 10.1016/j.cma.2019.03.039

Volume: 351

Issue:

First Page #: 20

Date Submitted: 7/9/19 12:00AM

Date Published: 7/1/19 12:00AM

Publication Location:

Article Title: Adaptive finite element simulations of waveguide configurations involving parallel 2D material sheets

Authors: Jung Heon Song, Matthias Maier, Mitchell Luskin

Keywords: Waveguide configurations; Time-harmonic Maxwell's equations; Adaptive finite-element methods; Surface plasmon-polariton

Abstract: We discuss analytically and numerically the propagation and energy transmission of electromagnetic waves caused by the coupling of surface plasmon polaritons (SPPs) between two spatially separated layers of 2D materials, such as graphene, at subwavelength distances. We construct an adaptive finite-element method to compute the ratio of energy transmitted within these waveguide structures reliably and efficiently. At its heart, the method is built upon a goal-oriented a posteriori error estimation with the dual-weighted residual method (DWR). Furthermore, we derive analytic solutions of the two-layer system, compare those to (known) single-layer configurations, and compare and validate our numerical findings by comparing numerical and analytical values for optimal spacing of the two-layer configuration. Additional aspects of our numerical treatment, such as local grid refinement, and the utilization of perfectly matched layers (PMLs) are examined in detail.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

Publication Type: Journal Article

Peer Reviewed: Y

Publication Status: 1-Published

Journal: Physical Review B

Publication Identifier Type: DOI

Publication Identifier: 10.1103/PhysRevB.99.201405

Volume: 99

Issue: 20

First Page #:

Date Submitted: 7/9/19 12:00AM

Date Published: 5/1/19 5:00AM

Publication Location:

Article Title: Switchable and unidirectional plasmonic beacons in hyperbolic two-dimensional materials

Authors: Andrei Nemilentsau, Tobias Stauber, Guillermo Gómez-Santos, Mitchell Luskin, Tony Low

Keywords: unidirectional, plasmon, hyperbolic, two-dimensional

Abstract: In hyperbolic two-dimensional (2D) materials, energy is channeled into their deep subwavelength polaritonic modes via four narrow beams. Here, we consider the launching of surface polaritons in hyperbolic 2D materials and demonstrate that efficient unidirectional excitation is possible with an elliptically polarized electric dipole, with the optimal choice of dipole ellipticity depending on the materials' optical constants. The selection rules afforded by the choice of dipole polarization allow turning off up to two beams, and even three if the dipole is placed close to an edge. This makes the dipole a directionally switchable beacon for the launching of subdiffractive polaritonic beams, a potential logical gate. We develop an analytical approximation of the excitation process which describes well the results of the numerical simulations and affords a simple physical interpretation.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: Physical Review Letters
Publication Identifier Type: DOI **Publication Identifier:** 10.1103/PhysRevLett.122.146801
Volume: 122 **Issue:** 14 **First Page #:**
Date Submitted: 7/9/19 12:00AM **Date Published:** 4/1/19 5:00AM
Publication Location:

Article Title: Tunneling Spectroscopy of Quantum Hall States in Bilayer Graphene

Authors: Ke Wang, Achim Harzheim, Takashi Taniguchi, Kenji Watanabei, Ji Ung Lee, Philip Kim

Keywords: Tunneling Spectroscopy, Quantum Hall States, Bilayer Graphene

Abstract: We report tunneling transport in spatially controlled networks of quantum Hall (QH) edge states in bilayer graphene. By manipulating the separation, location, and spatial span of QH edge states via gate-defined electrostatics, we observe resonant tunneling between copropagating QH states across incompressible strips. Employing spectroscopic tunneling measurements and an analytical model, we characterize the energy gap, width, density of states, and compressibility of the QH edge states with high precision and sensitivity within the same device. The capability to engineer the QH edge network also provides an opportunity to build future quantum electronic devices with electrostatic manipulation of QH edge states, supported by rich underlying physics.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: Nanotechnology
Publication Identifier Type: DOI **Publication Identifier:** 10.1088/1361-6528/aae7df
Volume: 30 **Issue:** 2 **First Page #:** 025201
Date Submitted: 7/9/19 12:00AM **Date Published:** 1/1/19 5:00AM
Publication Location:

Article Title: High performance tunnel field effect transistors based on in-plane transition metal dichalcogenide heterojunctions

Authors: Jean Choukroun, Marco Pala, Shiang Fang, Efthimios Kaxiras, Philippe Dollfus

Keywords: TMDC, FET

Abstract: In-plane heterojunction tunnel field effect transistors based on monolayer transition metal dichalcogenides are studied by means of self-consistent non-equilibrium Green's functions simulations and an atomistic tight-binding Hamiltonian. We start by comparing several heterojunctions before focusing on the most promising ones, i.e. WTe₂-MoS₂ and MoTe₂-MoS₂. The scalability of those devices as a function of channel length is studied, and the influence of backgate voltages on device performance is analyzed. Our results indicate that, by fine-tuning the design parameters, those devices can yield extremely low subthreshold swings (<5 mV/decade) and I_{ON}/I_{OFF} ratios higher than 10⁸ at a supply voltage of 0.3 V, making them ideal for ultra-low power consumption.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article

Peer Reviewed: Y

Publication Status: 1-Published

Journal: Nature

Publication Identifier Type: DOI

Publication Identifier: 10.1038/s41586-018-0807-6

Volume: 565

Issue: 7739

First Page #: 337

Date Submitted: 7/9/19 12:00AM

Date Published: 12/1/18 5:00AM

Publication Location:

Article Title: Observation of the nonlinear Hall effect under time-reversal-symmetric conditions

Authors: Qiong Ma, Su-Yang Xu, Huitao Shen, David MacNeill, Valla Fatemi, Tay-Rong Chang, Andrés M. Mier \

Keywords: TMDC, Hall Effect, Berry Curvature

Abstract: The electrical Hall effect is the production, upon the application of an electric field, of a transverse voltage under an out-of-plane magnetic field. Studies of the Hall effect have led to important breakthroughs, including the discoveries of Berry curvature and topological Chern invariants. The internal magnetization of magnets means that the electrical Hall effect can occur in the absence of an external magnetic field; this 'anomalous' Hall effect is important for the study of quantum magnets. The electrical Hall effect has rarely been studied in non-magnetic materials without external magnetic fields, owing to the constraint of time-reversal symmetry. However, only in the linear response regime—when the Hall voltage is linearly proportional to the external electric field—does the Hall effect identically vanish as a result of time-reversal symmetry; the Hall effect in the nonlinear response regime is not subject to such symmetry constraints. Here we report observations of the nonlin

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

Publication Type: Journal Article

Peer Reviewed: Y

Publication Status: 1-Published

Journal: Nature Communications

Publication Identifier Type: DOI

Publication Identifier: 10.1038/s41467-019-08904-9

Volume: 10

Issue: 1

First Page #:

Date Submitted: 7/9/19 12:00AM

Date Published: 3/1/19 5:00AM

Publication Location:

Article Title: Atomic electrostatic maps of 1D channels in 2D semiconductors using 4D scanning transmission electron microscopy

Authors: Shiang Fang, Yi Wen, Christopher S. Allen, Colin Ophus, Grace G. D. Han, Angus I. Kirkland, Efthimios

Keywords: TMDC, DFT, defects

Abstract: Defects in materials give rise to fluctuations in electrostatic fields that reflect the local charge density, but imaging this with single atom sensitivity is challenging. However, if possible, this provides information about the energetics of adatom binding, localized conduction channels, molecular functionality and their relationship to individual bonds. Here, ultrastable electron-optics are combined with a high-speed 2D electron detector to map electrostatic fields around individual atoms in 2D monolayers using 4D scanning transmission electron microscopy. Simultaneous imaging of the electric field, phase, annular dark field and the total charge in 2D MoS₂ and WS₂ is demonstrated for pristine areas and regions with 1D wires. The in-gap states in sulphur line vacancies cause 1D electron-rich channels that are mapped experimentally and confirmed using density functional theory calculations. We show how electrostatic fields are sensitive in defective areas to changes of atomic bonding

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: Physical Review B
Publication Identifier Type: DOI **Publication Identifier:** 10.1103/PhysRevB.98.235404
Volume: 98 **Issue:** 23 **First Page #:**
Date Submitted: 7/9/19 12:00AM **Date Published:** 12/1/18 5:00AM
Publication Location:

Article Title: Dihedral-angle-corrected registry-dependent interlayer potential for multilayer graphene structures

Authors: Mingjian Wen, Stephen Carr, Shiang Fang, Efthimios Kaxiras, Ellad B. Tadmor

Keywords: Graphene, Interatomic potential

Abstract: The structural relaxation of multilayer graphene is essential in describing the interesting electronic properties induced by intentional misalignment of successive layers, including the recently reported superconductivity in twisted bilayer graphene. This is difficult to accomplish without an accurate interatomic potential. Here, we present a new, registry-dependent Kolmogorov-Crespi-type interatomic potential to model interlayer interactions in multilayer graphene structures. It consists of two parts, representing attractive interaction due to dispersion and repulsive interaction due to anisotropic overlap of electronic orbitals. An important new feature is a dihedral-angle-dependent term that is added to the repulsive part to describe correctly several distinct stacking states that the original Kolmogorov-Crespi potential cannot distinguish. We refer to the new model as the dihedral-angle-corrected registry-dependent interlayer potential (DRIP). Computations for several test problems

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: The Journal of Physical Chemistry C
Publication Identifier Type: DOI **Publication Identifier:** 10.1021/acs.jpcc.8b07548
Volume: 122 **Issue:** 43 **First Page #:** 24535
Date Submitted: 7/10/19 12:00AM **Date Published:** 10/1/18 4:00AM
Publication Location:

Article Title: Lithium Intercalation in Graphene–MoS

Authors: Daniel T. Larson, Ioanna Fampiou, Gunn Kim, Efthimios Kaxiras

Keywords: Graphene, TMDC, Intercalation

Abstract: Two-dimensional (2D) heterostructures are interesting candidates for efficient energy storage devices due to their high carrier capacity by reversible intercalation. We employ here density functional theory calculations to investigate the structural and electronic properties of lithium-intercalated graphene/molybdenum disulfide (Gr/MoS₂) heterostructures. We explore the extent to which Li intercalates at the interface formed between graphene (Gr) and molybdenum disulfide (MoS₂) layers by considering the adsorption and diffusion of Li atoms, the energetic stability, and the changes in the structural morphology of MoS₂. We investigate the corresponding electronic structure and charge distribution within the heterostructure at varying concentrations of Li. Our results indicate that the maximum energetically allowed ratio of Li to Mo (Li to C) is 1:1 (1:3) for both the 2H and 1T⁺ phases of MoS₂. This is double the Li concentration allowed in graphene bilayers. We find that there is 60% more

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article

Peer Reviewed: Y

Publication Status: 1-Published

Journal: Nano Letters

Publication Identifier Type: DOI

Publication Identifier: 10.1021/acs.nanolett.8b03005

Volume: 18

Issue: 9

First Page #: 6057

Date Submitted: 7/10/19 12:00AM

Date Published: 8/1/18 4:00AM

Publication Location:

Article Title: New Pathway for Hot Electron Relaxation in Two-Dimensional Heterostructures

Authors: Jin Zhang, Hao Hong, Jia Zhang, Huixia Fu, Peiwei You, Johannes Lischner, Kaihui Liu, Efthimios Kaxiras

Keywords: TMDC, TD-DFT

Abstract: Two-dimensional (2D) heterostructures composed of transition-metal dichalcogenide atomic layers are the new frontier for novel optoelectronic and photovoltaic device applications. Some key properties that make these materials appealing, yet are not well understood, are ultrafast hole/electron dynamics, interlayer energy transfer and the formation of interlayer hot excitons. Here, we study photoexcited electron/hole dynamics in a representative heterostructure, the MoS₂/WSe₂ interface, which exhibits type II band alignment. Employing time-dependent density functional theory in the time domain, we observe ultrafast charge dynamics with lifetimes of tens to hundreds of femtoseconds. Most importantly, we report the discovery of an interfacial pathway in 2D heterostructures for the relaxation of photoexcited hot electrons through interlayer hopping, which is significantly faster than intralayer relaxation. This finding is of particular importance for understanding many experimentally observed

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

Publication Type: Journal Article

Peer Reviewed: Y

Publication Status: 1-Published

Journal: EPL (Europhysics Letters)

Publication Identifier Type: DOI

Publication Identifier: 10.1209/0295-5075/122/27003

Volume: 122

Issue: 2

First Page #: 27003

Date Submitted: 7/10/19 12:00AM

Date Published: 6/1/18 4:00AM

Publication Location:

Article Title: Emergence and dynamical properties of stochastic branching in the electronic flows of disordered Dirac solids

Authors: Marios Mattheakis, G. P. Tsironis, Efthimios Kaxiras

Keywords: Graphene, Disorder

Abstract: Graphene as well as more generally Dirac solids constitute two-dimensional materials where the electronic flow is ultra-relativistic. When a Dirac solid is deposited on a different substrate surface with roughness, a local random potential develops through an inhomogeneous charge impurity distribution. This external potential affects profoundly the charge flow and induces a chaotic pattern of current branches that develops through focusing and defocusing effects produced by the randomness of the surface. An additional bias voltage may be used to tune the branching pattern of the charge carrier currents. We employ analytical and numerical techniques in order to investigate the onset and the statistical properties of carrier branches in Dirac solids. We find a specific scaling-type relationship that connects the physical scale for the occurrence of branches with the characteristic medium properties, such as disorder and bias field. We use numerics to test and verify the theoretical predictions.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article

Peer Reviewed: Y

Publication Status: 1-Published

Journal: Physical Review B

Publication Identifier Type: DOI

Publication Identifier: 10.1103/PhysRevB.97.195435

Volume: 97

Issue: 19

First Page #:

Date Submitted: 7/9/19 12:00AM

Date Published: 5/1/18 12:00AM

Publication Location:

Article Title: Quantum plasmons with optical-range frequencies in doped few-layer graphene

Authors: Sharmila N. Shirodkar, Marios Mattheakis, Paul Cazeaux, Prineha Narang, Marin Soljačić, Efthimios K

Keywords: plasmons, graphene, dopants

Abstract: Although plasmon modes exist in doped graphene, the limited range of doping achieved by gating restricts the plasmon frequencies to a range that does not include the visible and infrared. Here we show, through the use of first-principles calculations, that the high levels of doping achieved by lithium intercalation in bilayer and trilayer graphene shift the plasmon frequencies into the visible range. To obtain physically meaningful results, we introduce a correction of the effect of plasmon interaction across the vacuum separating periodic images of the doped graphene layers, consisting of transparent boundary conditions in the direction perpendicular to the layers; this represents a significant improvement over the exact Coulomb cutoff technique employed in earlier works. The resulting plasmon modes are due to local field effects and the nonlocal response of the material to external electromagnetic fields, requiring a fully quantum mechanical treatment. We describe the features of the

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

Publication Type: Journal Article

Peer Reviewed: Y

Publication Status: 1-Published

Journal: Journal of Physics Communications

Publication Identifier Type: DOI

Publication Identifier: 10.1088/2399-6528/aa90c8

Volume: 1

Issue: 4

First Page #: 045003

Date Submitted: 7/10/19 12:00AM

Date Published: 11/1/17 4:00AM

Publication Location:

Article Title: Manipulating polarized light with a planar slab of black phosphorus

Authors: Constantinos A Valagiannopoulos, Marios Mattheakis, Sharmila N Shirodkar, Efthimios Kaxiras

Keywords: Photonics, Black Phosphorus

Abstract: Wave polarization contains valuable information for electromagnetic signal processing; thus, the ability to manipulate it, can be extremely useful in modeling photonic devices. In this work, we propose designs comprised of one of the emerging and interesting media: black phosphorus (BP). Due to substantial in-plane anisotropy, a single slab of BP can be very efficient for controlling the polarization state of electromagnetic waves. We investigate BP slabs that filter the fields along one direction, or achieve polarization axis rotation, or convert linear polarization to circular. These slabs can be employed as components in numerous mid-IR integrated structures.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: Physical Review B
Publication Identifier Type: DOI **Publication Identifier:** 10.1103/PhysRevB.96.165436
Volume: 96 **Issue:** 16 **First Page #:**
Date Submitted: 7/10/19 12:00AM **Date Published:** 10/1/17 4:00AM
Publication Location:

Article Title: Nanoribbon edges of transition-metal dichalcogenides: Stability and electronic properties

Authors: Daphne Davelou, Georgios Kopidakis, Efthimios Kaxiras, Ioannis N. Remediakis

Keywords: TMDC, edge states

Abstract: Nanoribbons of molybdenum disulfide (MoS₂) are interesting one-dimensional (1D) nanostructures with intriguing electronic properties, consisting of a semiconducting bulk bounded by edges with metallic character. Edges of similar character can also be expected in other transition-metal dichalcogenide (TMDC) nanostructures. We report first-principles electronic structure calculations for the total energy and the band structure of four representative TMDCs, MoS₂ , MoSe₂ , WS₂ , WSe₂ , in various 1D nanoribbon configurations. We compare the thermodynamic stability and the electronic structure of the 2D bulk and 35 different quasi-1D nanoribbons for each of the four materials. In each case, we consider the reconstructions of the zigzag metal-terminated edge by adding different amounts of chalcogen adatoms. The 1D structures we investigated have positive edge energies when the chalcogen chemical potential is close to the energy of the bulk chalcogen phase, and negative edge energies f

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: Physical Review B
Publication Identifier Type: DOI **Publication Identifier:** 10.1103/PhysRevB.95.235310
Volume: 95 **Issue:** 23 **First Page #:**
Date Submitted: 7/10/19 12:00AM **Date Published:** 6/1/17 12:00AM
Publication Location:

Article Title: Accurate formation energies of charged defects in solids: A systematic approach

Authors: Dmitry Vinichenko, Mehmet Gokhan Sensoy, Cynthia M. Friend, Efthimios Kaxiras

Keywords: Defects, TMDC

Abstract: Defects on surfaces of semiconductors have a strong effect on their reactivity and catalytic properties. The concentration of different charge states of defects is determined by their formation energies. First-principles calculations are an important tool for computing defect formation energies and for studying the microscopic environment of the defect. The main problem associated with the widely used supercell method in these calculations is the error in the electrostatic energy, which is especially pronounced in calculations that involve surface slabs and two-dimensional materials. We present an internally consistent approach for calculating defect formation energies in inhomogeneous and anisotropic dielectric environments and demonstrate its applicability to the cases of the positively charged Cl vacancy on the NaCl (100) surface and the negatively charged S vacancy in monolayer MoS₂.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: Physical Review B
Publication Identifier Type: DOI **Publication Identifier:** 10.1103/PhysRevB.94.241303
Volume: 94 **Issue:** 24 **First Page #:**
Date Submitted: 7/10/19 12:00AM **Date Published:** 12/1/16 5:00AM
Publication Location:

Article Title: Electric field tuning of band offsets in transition metal dichalcogenides

Authors: Dennis Huang, Efthimios Kaxiras

Keywords: TMDC, DFT, Excitons

Abstract: We use first-principles calculations to investigate the band structure evolution of $W X_2 / Mo X_2$ ($X = S, Se$) heterobilayers under a perpendicular electric field. We characterize the extent to which the type II band alignment in these compounds can be tuned or inverted electrostatically. Our results demonstrate two effects of the stacking configuration. First, different stackings produce different net dipole moments, resulting in band offset variations that are larger than 0.1 eV. Second, based on symmetry constraints that depend on stacking, a perpendicular electric field may hybridize $W X_2$ and $Mo X_2$ bands that cross at the Brillouin zone corner K. Our results suggest that external electric fields can be used to tune the physics of intralayer and interlayer excitons in heterobilayers of transition metal dichalcogenides.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: Nano Letters
Publication Identifier Type: DOI **Publication Identifier:** 10.1021/acs.nanolett.6b03999
Volume: 16 **Issue:** 12 **First Page #:** 7798
Date Submitted: 7/10/19 12:00AM **Date Published:** 11/1/16 4:00AM
Publication Location:

Article Title: MoS₂ Field-Effect Transistor with Sub-10 nm Channel Length

Authors: Amirhasan Nourbakhsh, Ahmad Zubair, Redwan N. Sajjad, Amir Tavakkoli K. G., Wei Chen, Shiang Far

Keywords: TMDC, FET

Abstract: Atomically thin molybdenum disulfide (MoS₂) is an ideal semiconductor material for field-effect transistors (FETs) with sub-10 nm channel lengths. The high effective mass and large bandgap of MoS₂ minimize direct source-drain tunneling, while its atomically thin body maximizes the gate modulation efficiency in ultrashort-channel transistors. However, no experimental study to date has approached the sub-10 nm scale due to the multiple challenges related to nanofabrication at this length scale and the high contact resistance traditionally observed in MoS₂ transistors. Here, using the semiconducting-to-metallic phase transition of MoS₂, we demonstrate sub-10 nm channel-length transistor fabrication by directed self-assembly patterning of mono- and trilayer MoS₂. This is done in a 7.5 nm half-pitch periodic chain of transistors where semiconducting (2H) MoS₂ channel regions are seamlessly connected to metallic-phase (1T') MoS₂ access and contact regions. The resulting 7.5 nm channel-length

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article

Peer Reviewed: Y

Publication Status: 1-Published

Journal: Physical Review B

Publication Identifier Type: DOI

Publication Identifier: 10.1103/PhysRevB.94.155310

Volume: 94

Issue: 15

First Page #:

Date Submitted: 7/9/19 12:00AM

Date Published: 10/1/16 12:00AM

Publication Location:

Article Title: Strain dependence of band gaps and exciton energies in pure and mixed transition-metal dichalcogenides

Authors: Rodrick Kuate Defo, Shiang Fang, Sharmila N. Shirodkar, Georgios A. Tritsarlis, Athanasios Dimoulas, E

Keywords: TMDC, strain, DFT

Abstract: The ability to fabricate 2D device architectures with desired properties, based on stacking of weakly (van der Waals) interacting atomically thin layers, is quickly becoming reality. In order to design ever more complex devices of this type, it is crucial to know the precise strain and composition dependence of the layers' electronic and optical properties. Here, we present a theoretical study of these dependences for monolayers with compositions varying from pure MX_2 to the mixed MXY , where $M = Mo, W$ and $X, Y = S, Se$. We employ both density-functional-theory and GW calculations, as well as values of the exciton binding energies based on a self-consistent treatment of dielectric properties, to obtain the band gaps that correspond to optical or transport measurements; we find reasonable agreement with reported experimental values for the unstrained monolayers. Our predictions for the strain-dependent electronic properties should be a useful guide in the effort to design heterost

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

Publication Type: Journal Article

Peer Reviewed: Y

Publication Status: 1-Published

Journal: Physical Review B

Publication Identifier Type: DOI

Publication Identifier: 10.1103/PhysRevB.94.201404

Volume: 94

Issue: 20

First Page #:

Date Submitted: 7/9/19 12:00AM

Date Published: 11/1/16 4:00AM

Publication Location:

Article Title: Epsilon-near-zero behavior from plasmonic Dirac point: Theory and realization using two-dimensional materials

Authors: Marios Mattheakis, Constantinos A. Valagiannopoulos, Efthimios Kaxiras

Keywords: Plasmonics, Photonics

Abstract: The electromagnetic response of a two-dimensional metal embedded in a periodic array of a dielectric host can give rise to a plasmonic Dirac point that emulates epsilon-near-zero (ENZ) behavior. This theoretical result is extremely sensitive to structural features like periodicity of the dielectric medium and thickness imperfections. We propose that such a device can actually be realized by using graphene as the two-dimensional metal and materials like the layered semiconducting transition-metal dichalcogenides or hexagonal boron nitride as the dielectric host. We propose a systematic approach, in terms of design characteristics, for constructing metamaterials with linear, elliptical, and hyperbolic dispersion relations which produce ENZ behavior, normal or negative diffraction.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article

Peer Reviewed: Y

Publication Status: 1-Published

Journal: Physical Review Letters

Publication Identifier Type: DOI

Publication Identifier: 10.1103/PhysRevLett.117.116804

Volume: 117 Issue: 11

First Page #:

Date Submitted: 7/9/19 12:00AM

Date Published: 9/1/16 4:00AM

Publication Location:

Article Title: Superlattice-Induced Insulating States and Valley-Protected Orbits in Twisted Bilayer Graphene

Authors: Y. Cao, J. Y. Luo, V. Fatemi, S. Fang, J. D. Sanchez-Yamagishi, K. Watanabe, T. Taniguchi, E. Kaxiras

Keywords: graphene, twist

Abstract: Twisted bilayer graphene (TBLG) is one of the simplest van der Waals heterostructures, yet it yields a complex electronic system with intricate interplay between moiré physics and interlayer hybridization effects. We report on electronic transport measurements of high mobility small angle TBLG devices showing clear evidence for insulating states at the superlattice band edges, with thermal activation gaps several times larger than theoretically predicted. Moreover, Shubnikov–de Haas oscillations and tight binding calculations reveal that the band structure consists of two intersecting Fermi contours whose crossing points are effectively unhybridized. We attribute this to exponentially suppressed interlayer hopping amplitudes for momentum transfers larger than the moiré wave vector.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

Publication Type: Journal Article

Peer Reviewed: Y

Publication Status: 1-Published

Journal: Physical Review Letters

Publication Identifier Type: DOI

Publication Identifier: 10.1103/PhysRevLett.117.056804

Volume: 117 Issue: 5

First Page #:

Date Submitted: 7/9/19 12:00AM

Date Published: 7/1/16 4:00AM

Publication Location:

Article Title: High-Temperature Quantum Anomalous Hall Effect in n?p Codoped Topological Insulators

Authors: Shifei Qi, Zhenhua Qiao, Xinzhou Deng, Ekin D. Cubuk, Hua Chen, Wenguang Zhu, Efthimios Kaxiras, :

Keywords: Hall Effect, Topological Insulator

Abstract: The quantum anomalous Hall effect (QAHE) is a fundamental quantum transport phenomenon that manifests as a quantized transverse conductance in response to a longitudinally applied electric field in the absence of an external magnetic field, and it promises to have immense application potential in future dissipationless quantum electronics. Here, we present a novel kinetic pathway to realize the QAHE at high temperatures by n ? p codoping of three-dimensional topological insulators. We provide a proof-of-principle numerical demonstration of this approach using vanadium-iodine (V-I) codoped Sb₂Te₃ and demonstrate that, strikingly, even at low concentrations of ~2 % V and ~1 % I, the system exhibits a quantized Hall conductance, the telltale hallmark of QAHE, at temperatures of at least ~50 K, which is 3 orders of magnitude higher than the typical temperatures at which it has been realized to date. The underlying physical factor enabling this dramatic improvement is tied to the lar

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article

Peer Reviewed: Y

Publication Status: 1-Published

Journal: Nano Letters

Publication Identifier Type: DOI

Publication Identifier: 10.1021/acs.nanolett.6b01163

Volume: 16

Issue: 7

First Page #: 4224

Date Submitted: 7/9/19 12:00AM

Date Published: 6/1/16 4:00AM

Publication Location:

Article Title: Dumbbell Defects in FeSe Films: A Scanning Tunneling Microscopy and First-Principles Investigation

Authors: Dennis Huang, Tatiana A. Webb, Can-Li Song, Cui-Zu Chang, Jagadeesh S. Moodera, Efthimios Kaxira

Keywords: STM, DFT, FeSe, defect

Abstract: The properties of iron-based superconductors (Fe-SCs) can be varied dramatically with the introduction of dopants and atomic defects. As a pressing example, FeSe, parent phase of the highest-T_c Fe-SC, exhibits prevalent defects with atomic-scale “dumbbell” signatures as imaged by scanning tunneling microscopy (STM). These defects spoil superconductivity when their concentration exceeds 2.5%. Resolving their chemical identity is a prerequisite to applications such as nanoscale patterning of superconducting/nonsuperconducting regions in FeSe as well as fundamental questions such as the mechanism of superconductivity and the path by which the defects destroy it. We use STM and density functional theory to characterize and identify the dumbbell defects. In contrast to previous speculations about Se adsorbates or substitutions, we find that an Fe-site vacancy is the most energetically favorable defect in Se-rich conditions and reproduces our observed STM signature. Our calculations shed light

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

Publication Type: Journal Article

Peer Reviewed: Y

Publication Status: 1-Published

Journal: Physical Review B

Publication Identifier Type: DOI

Publication Identifier: 10.1103/PhysRevB.99.094431

Volume: 99

Issue: 9

First Page #: 094431

Date Submitted: 8/13/19 12:00AM

Date Published: 3/1/19 5:00AM

Publication Location:

Article Title: Quantum spin transfer torque induced nonclassical magnetization dynamics and electron-magnetization entanglement

Authors: Priyanka Mondal, Utkarsh Bajpai, Marko D. Petrovic, Petr Plechac, Branislav K. Nikolic

Keywords: quantum spin transfer, electron-magnetization entanglement

Abstract: The standard spin transfer torque (STT)—where spin-polarized current drives the dynamics of magnetization viewed as a classical vector—requires noncollinearity between electron spins carried by the current and magnetization of a ferromagnetic layer. However, recent experiments [A. Zholud et al., Phys. Rev. Lett. 119, 257201 (2017)] observing magnetization dynamics in spin valves at cryogenic temperatures, even when electron spin is collinear to magnetization, point at overlooked quantum effects in STT that can lead to highly nonclassical magnetization states. Using quantum many-body treatment, where an electron injected as a spin-polarized wave packet interacts with local spins comprising the anisotropic quantum Heisenberg ferromagnetic chain, we define quantum STT as any time evolution of local spins due to an initial many-body quantum state not being an eigenstate of an electron+local-spins composite system.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article Peer Reviewed: N **Publication Status:** 5-Submitted
Journal: Phys. Rev. Lett
Publication Identifier Type: **Publication Identifier:**
Volume: **Issue:** **First Page #:**
Date Submitted: 8/13/19 12:00AM **Date Published:** 8/9/19 4:00AM
Publication Location:

Article Title: Annihilation of magnetic domain walls induced spin wave burst intertwined with spin pumping: A multiscale time-dependent-quantum-transport/classical-micromagnetics approach

Authors: M. Petrovic, P. Plechac, B. K. Nikolic

Keywords: quantum spin transport, spin waves, coupling quantum-classical dynamics

Abstract: Motivated by recent experiments [S. Woo, Nat. Phys. 13, 448 (2017)] on magnetic-field-driven annihilation of magnetic domain walls (DWs) and the ensuing burst of spin waves (SWs) of very short wavelengths, we predict that this is also accompanied by pumping of electronic spin current whose time-dependence encodes information about the moment of the burst. The pumping occurs in the absence of bias voltage, and it is due to time-dependent fields introduced into the quantum Hamiltonian of conduction electrons by the classical dynamics of localized magnetic moments comprising the domains. The pumped currents carry spin-polarized electrons which, in turn, exert spin-transfer torque on localized magnetic moments, so that the self-consistent nonequilibrium state of the whole system cannot be captured by purely classical micromagnetic simulations where quantum time evolution of electrons is absent. Instead, we employ recently developed quantum-transport/classical-micromagnetics formalism.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: Communications in Mathematical Sciences
Publication Identifier Type: DOI **Publication Identifier:** CMS.2019.v17.n8.a5
Volume: 17 **Issue:** 8 **First Page #:** 2191
Date Submitted: 8/16/20 12:00AM **Date Published:** 7/8/19 12:00AM
Publication Location:

Article Title: The classical limit of quantum observables in the conservation laws

Authors: P. Plechac, M. Sandberg, A. Szepessy

Keywords: conservation laws, stress tensor, heat flux, molecular dynamics, Weyl quantization

Abstract: In the classical work by Irving and Zwanzig [Irving J.H. and Zwanzig R.W., J. Chem. Phys. 19 (1951), 1173-1180] it has been shown that quantum observables for macroscopic density, momentum and energy satisfy the conservation laws of fluid dynamics. This work derives the corresponding classical molecular dynamics limit by extending Irving and Zwanzig's result to matrix-valued potentials for a general quantum particle system. The matrix formulation provides the semi-classical limit of the quantum observables in the conservation laws, also in the case where the temperature is large compared to the electron eigenvalue gaps. The classical limit of the quantum observables in the conservation laws is useful in order to determine the constitutive relations for the stress tensor and the heat flux by molecular dynamics simulations. The main new steps to obtain the molecular dynamics limit is to: (i) approximate the dynamics of quantum observables accurately by classical dynamics.

Distribution Statement: 2-Distribution Limited to U.S. Government agencies only; report contains proprietary info

Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article

Peer Reviewed: Y

Publication Status: 1-Published

Journal: physica status solidi (b)

Publication Identifier Type: DOI

Publication Identifier: 10.1002/pssb.201800191

Volume: 255

Issue: 12

First Page #: 1800191

Date Submitted: 8/14/19 12:00AM

Date Published: 8/1/18 5:00AM

Publication Location:

Article Title: Measuring the Local Twist Angle and Layer Arrangement in Van der Waals Heterostructures

Authors: Tobias A. de Jong, Johannes Jobst, Hyobin Yoo, Eugene E. Krasovskii, Philip Kim, Sense Jan van der I

Keywords: Van der Waals

Abstract: The properties of Van der Waals (VdW) heterostructures are determined by the twist angle and the interface between adjacent layers as well as their polytype and stacking. Here, the use of spectroscopic low energy electron microscopy (LEEM) and micro low energy electron diffraction (mLEED) methods to measure these properties locally is described. The authors present results on a MoS₂/hBN heterostructure, but the methods are applicable to other materials. Diffraction spot analysis is used to assess the benefits of using hBN as a substrate. In addition, by making use of the broken rotational symmetry of the lattice, the cleaving history of the MoS₂ flake is determined, that is, which layer stems from where in the bulk.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

Publication Type: Journal Article

Peer Reviewed: Y

Publication Status: 1-Published

Journal: Archive for Rational Mechanics and Analysis

Publication Identifier Type: DOI

Publication Identifier: 10.1007/s00205-019-01444-y

Volume: 235

Issue: 2

First Page #: 1289

Date Submitted: 7/25/20 12:00AM

Date Published: 8/1/19 5:00AM

Publication Location:

Article Title: Energy Minimization of Two Dimensional Incommensurate Heterostructures

Authors: Paul Cazeaux, Mitchell Luskin, Daniel Massatt

Keywords: energy minimization, incommensurate

Abstract: We derive and analyze a novel approach for modeling and computing the mechanical relaxation of incommensurate two dimensional heterostructures. Our approach parametrizes the relaxation pattern by the compact local configuration space rather than real space, thus bypassing the need for the standard supercell approximation and giving a true aperiodic atomistic configuration. Our model extends the computationally accessible regime of weakly coupled bilayers with similar orientations or lattice spacing, for example materials with a small relative twist where the widely studied large-scale moiré patterns arise (Kim et al. in Proc Natl Acad Sci 114:3364–3369, 2017; Yoo et al. in Atomic and electronic reconstruction at van derWaals interface in twisted bilayer graphene, NatMater 18:448–453, 2019). Our model also makes possible the simulation of multi-layers for which no interlayer empirical atomistic potential exists, such as those composed of MoS₂ layers, and more generally makes possible t

Distribution Statement: 2-Distribution Limited to U.S. Government agencies only; report contains proprietary info

Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Proceedings of the Royal Society A Mathematical, Physical and Engineering Sciences

Publication Identifier Type: DOI

Publication Identifier: 10.1098/rspa.2019.0220

Volume: 475

Issue: 2230

First Page #: 20190220

Date Submitted: 7/25/20 12:00AM

Date Published: 10/1/19 5:00AM

Publication Location:

Article Title: Homogenization of plasmonic crystals; seeking the epsilon-near-zero effect

Authors: M. Maier, M. Mattheakis, E. Kaxiras, M. Luskin, D. Margetis

Keywords: homogenization, plasmon

Abstract: We derive and analyze a novel approach for modeling and computing the mechanical relaxation of incommensurate two dimensional heterostructures. Our approach parametrizes the relaxation pattern by the compact local configuration space rather than real space, thus bypassing the need for the standard supercell approximation and giving a true aperiodic atomistic configuration. Our model extends the computationally accessible regime of weakly coupled bilayers with similar orientations or lattice spacing, for example materials with a small relative twist where the widely studied large-scale moiré patterns arise (Kim et al. in Proc Natl Acad Sci 114:3364–3369, 2017; Yoo et al. in Atomic and electronic reconstruction at van derWaals interface in twisted bilayer graphene, NatMater 18:448–453, 2019). Our model also makes possible the simulation of multi-layers for which no interlayer empirical atomistic potential exists, such as those composed of MoS2 layers, and more generally makes possible t

Distribution Statement: 2-Distribution Limited to U.S. Government agencies only; report contains proprietary info

Acknowledged Federal Support: Y

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Journal of Physics A Mathematical and Theoretical

Publication Identifier Type: DOI

Publication Identifier: 10.1088/1751-8121/ab5ff9

Volume: 53

Issue: 5

First Page #: 055201

Date Submitted: 7/25/20 12:00AM

Date Published: 2/1/20 6:00AM

Publication Location:

Article Title: Nonretarded edge plasmon-polaritons in anisotropic two-dimensional materials

Authors: Dionisios Margetis, Matthias Maier, Tobias Stauber, Tony Low, Mitchell Luskin

Keywords: edge plasmon

Abstract: We derive and analyze a novel approach for modeling and computing the mechanical relaxation of incommensurate two dimensional heterostructures. Our approach parametrizes the relaxation pattern by the compact local configuration space rather than real space, thus bypassing the need for the standard supercell approximation and giving a true aperiodic atomistic configuration. Our model extends the computationally accessible regime of weakly coupled bilayers with similar orientations or lattice spacing, for example materials with a small relative twist where the widely studied large-scale moiré patterns arise (Kim et al. in Proc Natl Acad Sci 114:3364–3369, 2017; Yoo et al. in Atomic and electronic reconstruction at van derWaals interface in twisted bilayer graphene, NatMater 18:448–453, 2019). Our model also makes possible the simulation of multi-layers for which no interlayer empirical atomistic potential exists, such as those composed of MoS2 layers, and more generally makes possible t

Distribution Statement: 2-Distribution Limited to U.S. Government agencies only; report contains proprietary info

Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: The European Physical Journal B

Publication Identifier Type: DOI

Publication Identifier: 10.1140/epjb/e2020-100518-7

Volume: 93

Issue: 4

First Page #:

Date Submitted: 7/25/20 12:00AM

Date Published: 4/1/20 12:00AM

Publication Location:

Article Title: Efficient computation of Kubo conductivity for incommensurate 2D heterostructures

Authors: Daniel Massatt, Stephen Carr, Mitchell Luskin

Keywords: Kubo conductivity

Abstract: We introduce a numerical method for computing conductivity via the Kubo formula for incommensurate 2D bilayer heterostructures using a tight-binding framework. We begin by deriving the momentum space formulation and Kubo formula from the real space tight-binding model using the appropriate Bloch transformation operator. We further discuss the resulting algorithm along with its convergence rate and computational cost in terms of model parameters such as relaxation time and temperature. In particular, we show that for low frequencies, low temperature, and long relaxation times conductivity can be computed very efficiently using the momentum space algorithm for a wide class of materials. We then showcase our method by computing conductivity for twisted bilayer graphene (tBLG) for small twist angles.

Distribution Statement: 2-Distribution Limited to U.S. Government agencies only; report contains proprietary info
Acknowledged Federal Support: Y

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Nature Reviews Materials

Publication Identifier Type: DOI

Publication Identifier: <https://doi.org/10.1038/s41578-020-0214-0>

Volume:

Issue:

First Page #:

Date Submitted: 8/1/20 12:00AM

Date Published: 7/22/20 2:00AM

Publication Location:

Article Title: Electronic-structure methods for twisted moiré layers

Authors: Stephen Carr, Shiang Fang, and Efthimios Kaxiras

Keywords: electronic structure, twisted bilayers, two-dimensional materials

Abstract: When single layers of 2D materials are stacked on top of one another with a small twist in orientation, the resulting structure often involves incommensurate moiré patterns. In these patterns, the loss of angstrom-scale periodicity poses a significant theoretical challenge, and the new moiré length scale leads to emergent physical phenomena. The range of physics arising from twisted bilayers has led to significant advances that are shaping into a new field, twistronics. At the moiré scale, the large number of atoms in these systems can make their accurate simulation daunting, necessitating the development of efficient multiscale methods. In this Review, we summarize and compare such modelling methods — focusing in particular on density functional t

Distribution Statement: 2-Distribution Limited to U.S. Government agencies only; report contains proprietary info
Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article

Peer Reviewed: Y

Publication Status: 1-Published

Journal: Nano Letters

Publication Identifier Type: DOI

Publication Identifier: 10.1021/acs.nanolett.9b04979

Volume: 20

Issue: 5

First Page #: 3030

Date Submitted: 8/1/20 12:00AM

Date Published: 3/1/20 12:00AM

Publication Location:

Article Title: Ultraheavy and Ultrarelativistic Dirac Quasiparticles in Sandwiched Graphenes

Authors: Stephen Carr, Chenyuan Li, Ziyang Zhu, Efthimios Kaxiras, Subir Sachdev, Alexander Kruchkov

Keywords: Graphene, moire? superlattices, flat bands, ab initio calculations, lattice relaxation.

Abstract: Electrons in quantum materials exhibiting coexistence of dispersionless (flat) bands piercing dispersive (steep) bands give rise to strongly correlated phenomena and are associated with unconventional superconductivity. We show that in twisted sandwiched graphene (TSWG), a three-layer van der Waals heterostructure with a twisted middle layer, steep Dirac cones can coexist with dramatic band flattening at the same energy scale, if twisted by 1.5° . This phenomenon is not stable in the simplified continuum models. The key result of this Letter is that the flat bands become stable only as a consequence of lattice relaxation processes included in our atomistic calculations. Further on, external fields can change the relative energy offset between the Dirac cone vertex and the flat bands and enhance band hybridization, which could permit controlling correlated phases. Our work establishes twisted sandwiched graphene as a new platform for research into strongly interacting two-dimensional

Distribution Statement: 2-Distribution Limited to U.S. Government agencies only; report contains proprietary info
Acknowledged Federal Support: Y

Publication Type: Journal Article

Peer Reviewed: Y

Publication Status: 1-Published

Journal: Physical Review B

Publication Identifier Type: DOI

Publication Identifier: 10.1103/PhysRevB.101.224107

Volume: 101

Issue: 22

First Page #:

Date Submitted: 8/1/20 12:00AM

Date Published: 6/1/20 4:00AM

Publication Location:

Article Title: Modeling mechanical relaxation in incommensurate trilayer van der Waals heterostructures

Authors: Ziyang Zhu, Paul Cazeaux, Mitchell Luskin, Efthimios Kaxiras

Keywords: mechanical deformation, two-dimensional systems

Abstract: The incommensurate stacking of multilayered two-dimensional materials is a challenging problem from a theoretical perspective and an intriguing avenue for manipulating their physical properties. Here we present a multiscale model to obtain the mechanical relaxation pattern of twisted trilayer van der Waals (vdW) heterostructures with two independent twist angles, a generally incommensurate system without a supercell description. We adopt the configuration space as a natural description of such incommensurate layered materials, based on the local environment of atomic positions, bypassing the need for commensurate approximations. To obtain the relaxation pattern, we perform energy minimization with respect to the relaxation displacement vectors. We use a continuum model in combination with the generalized stacking fault energy to describe the interlayer coupling, obtained from first-principles calculations based on density functional theory. We show that the relaxation patterns of twist

Distribution Statement: 2-Distribution Limited to U.S. Government agencies only; report contains proprietary info
Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Journal of Chemical Theory and Computation

Publication Identifier Type: DOI

Publication Identifier: 10.1021/acs.jctc.9b00617

Volume: 15

Issue: 11

First Page #: 6036

Date Submitted: 8/1/20 12:00AM

Date Published: 9/1/19 4:00AM

Publication Location:

Article Title: Imaginary-Time Time-Dependent Density Functional Theory and Its Application for Robust Convergence of Electronic States

Authors: Cedric Flamant, Grigory Kolesov, Efstratios Manousakis, Efthimios Kaxiras

Keywords: time-dependent density functional theory

Abstract: Reliable and robust convergence to the electronic ground state within density functional theory (DFT) Kohn–Sham (KS) calculations remains a thorny issue in many systems of interest. In such cases, charge sloshing can delay or completely hinder the convergence. Here, we use an approach based on transforming the time-dependent DFT equations to imaginary time, followed by imaginary-time evolution, as a reliable alternative to the self-consistent field (SCF) procedure for determining the KS ground state. We discuss the theoretical and technical aspects of this approach and show that the KS ground state should be expected to be the long-imaginary-time output of the evolution, independent of the exchange-correlation functional or the level of theory used to simulate the system. By maintaining self-consistency between the single-particle wave functions (orbitals) and the electronic density throughout the determination of the stationary state, our method avoids the typical difficulties encountered

Distribution Statement: 2-Distribution Limited to U.S. Government agencies only; report contains proprietary information
Acknowledged Federal Support: Y

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Physical Review Research

Publication Identifier Type: DOI

Publication Identifier: 10.1103/PhysRevResearch.1.033072

Volume: 1

Issue: 3

First Page #:

Date Submitted: 8/1/20 12:00AM

Date Published: 11/1/19 4:00AM

Publication Location:

Article Title: Derivation of Wannier orbitals and minimal-basis tight-binding Hamiltonians for twisted bilayer graphene; First-principles approach

Authors: Stephen Carr, Shiang Fang, Hoi Chun Po, Ashvin Vishwanath, Efthimios Kaxiras

Keywords: Electronic structure, graphene, Wannier function methods

Abstract: Twisted bilayer graphene (TBLG) has emerged as an important platform for studying correlated phenomena, including unconventional superconductivity, in two-dimensional systems. The complexity of the atomic-scale structures in TBLG has made even the study of single-particle physics at low energies around the Fermi level, quite challenging. Our goal here is to provide a convenient and physically motivated picture of single-particle physics in TBLG using reduced models with the smallest possible number of localized orbitals. The reduced models exactly reproduce the low-energy bands of ab initio tight-binding models, including the effects of atomic relaxations. Furthermore, we obtain for the first time the corresponding Wannier orbitals that incorporate all symmetries of TBLG, which are also calculated as a function of angle, a requisite first step towards incorporating electron interaction effects. We construct eight-band and five-band models for the low-energy states for twist angles between

Distribution Statement: 2-Distribution Limited to U.S. Government agencies only; report contains proprietary information
Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Journal of Applied Physics

Publication Identifier Type: DOI

Publication Identifier: 10.1063/1.5099496

Volume: 126

Issue: 8

First Page #: 084303

Date Submitted: 8/1/20 12:00AM

Date Published: 8/1/19 4:00AM

Publication Location:

Article Title: First-principles study of coupled effect of ripplocations and S-vacancies in MoS

Authors: Georgios A. Tritsarlis, Mehmet Gökhan Ensoy, Sharmila N. Shirodkar, Efthimios Kaxiras

Keywords: Density functional theory, vacancies, two-dimensional materials

Abstract: Recent experiments have revealed ripplocations, atomic-scale ripplelike defects on samples of MoS₂ flakes. We use quantum mechanical calculations based on density functional theory to study the effect of ripplocations on the structural and electronic properties of single-layer MoS₂, and, in particular, the coupling between these extended defects and the most common defects in this material, S-vacancies. We find that the formation of neutral S-vacancies is energetically more favorable in the ripplocation. In addition, we demonstrate that ripplocations alone do not introduce electronic states into the intrinsic bandgap, in contrast to S-vacancies. We study the dependence of the induced gap states on the position of the defects in the ripplocation, which has implications for the experimental characterization of MoS₂ flakes and the engineering of quantum emitters in this material. Our specific findings collectively aim to provide insights into the electronic structure of experimentally relevant

Distribution Statement: 2-Distribution Limited to U.S. Government agencies only; report contains proprietary information

Acknowledged Federal Support: Y

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Nano Letters

Publication Identifier Type: DOI

Publication Identifier: acs.nanolett.9b04556

Volume: 20

Issue: 4

First Page #: 2288

Date Submitted: 8/16/20 12:00AM

Date Published: 3/3/20 12:00AM

Publication Location:

Article Title: Proximity Spin-Orbit Torque on a Two-Dimensional Magnet within van der Waals Heterostructure; Current-Driven Antiferromagnet-to-Ferromagnet Reversible Nonequilibrium Phase Transition in Bilayer CrI₃

Authors: Kapil Deb Doli, Marko D. Petrovic, Klaus Zollner, Petr Plechac, Jaroslav Fabian, and Branislav K. Nikolic

Keywords: orbit torque, 2D magnetic materials, van der Waals heterostructures, spintronics, first-principles quantum transport

Abstract: The recently discovered two-dimensional magnetic insulator CrI₃ is an intriguing case for basic research and spintronic applications since it is a ferromagnet in the bulk but an antiferromagnet in bilayer form, with its magnetic ordering amenable to external manipulations. Using the first-principles quantum transport approach, we predict that injecting unpolarized charge current parallel to the interface of the bilayer-CrI₃/monolayer-TaSe₂ van der Waals (vdW) heterostructure will induce spin-orbit torque and thereby drive the dynamics of magnetization on the first monolayer of CrI₃ in direct contact with TaSe₂. By combining the calculated complex angular dependence of spin-orbit torque with the Landau-Lifshitz-Gilbert equation for classical dynamics of magnetization, we demonstrate that current pulses can switch the direction of magnetization on the first monolayer to become parallel to that of the second monolayer, thereby converting CrI₃ from antiferromagnet to ferromagnet.

Distribution Statement: 2-Distribution Limited to U.S. Government agencies only; report contains proprietary information

Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article

Peer Reviewed: Y

Publication Status: 4-Under Review

Journal: Physical Review X

Publication Identifier Type:

Publication Identifier:

Volume:

Issue:

First Page #:

Date Submitted: 8/17/20 12:00AM

Date Published:

Publication Location:

Article Title: Spintronics meets density matrix renormalization group; Nonclassical magnetization reversal and entanglement growth due to current-pulse-driven quantum spin torque

Authors: M.D. Petrovic, P. Mondal, A.E. Feiguin, P. Plechac, B. K. Nikolic

Keywords: time-dependent density matrix renormalization group, non-equilibrium quantum dynamics, entanglement, spin-transfer torque

Abstract: Two-dimensional molybdenum disulfide (MoS₂) is a promising material for the next generation of switchable transistors and photodetectors. In order to perform large-scale molecular simulations of the mechanical and thermal behavior of MoS₂-based devices, an accurate interatomic potential is required. To this end, we have developed a Stillinger-Weber potential for monolayer MoS₂. The potential parameters are optimized to reproduce the geometry (bond lengths and bond angles) of MoS₂ in its equilibrium state and to match as closely as possible the forces acting on the atoms along a dynamical trajectory obtained from ab initio molecular dynamics.

Verification calculations indicate that the new potential accurately predicts important material properties including the strain dependence of the cohesive energy, the elastic constants, and the linear thermal expansion coefficient. The uncertainty in the potential parameters is determined using a Fisher information theory analysis.

Distribution Statement: 2-Distribution Limited to U.S. Government agencies only; report contains proprietary info
Acknowledged Federal Support: Y

Publication Type: Journal Article

Peer Reviewed: Y

Publication Status: 1-Published

Journal: npj Computational Materials

Publication Identifier Type: DOI

Publication Identifier: 10.1038/s41524-020-00390-8

Volume: 6

Issue: 1

First Page #:

Date Submitted: 8/28/20 12:00AM

Date Published: 8/1/20 5:00AM

Publication Location:

Article Title: Uncertainty quantification in molecular simulations with dropout neural network potentials

Authors: Mingjian Wen, Eyal B. Tadmor

Keywords: Uncertainty quantification, neural network potentials

Abstract: Machine learning interatomic potentials (IPs) can provide accuracy close to that of first-principles methods, such as density functional theory (DFT), at a fraction of the computational cost. This greatly extends the scope of accurate molecular simulations, providing opportunities for quantitative design of materials and devices on scales hitherto unreachable by DFT methods. However, machine learning IPs have a basic limitation in that they lack a physical model for the phenomena being predicted and therefore have unknown accuracy when extrapolating outside their training set. In this paper, we propose a class of Dropout Uncertainty Neural Network (DUNN) potentials that provide rigorous uncertainty estimates that can be understood from both Bayesian and frequentist statistics perspectives. As an example, we develop a DUNN potential for carbon and show how it can be used to predict uncertainty for static and dynamical properties, including stress and phonon dispersion in graphene.

Distribution Statement: 2-Distribution Limited to U.S. Government agencies only; report contains proprietary info
Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Physical Review B

Publication Identifier Type: DOI

Publication Identifier: 10.1103/PhysRevB.100.195419

Volume: 100 Issue: 19

First Page #:

Date Submitted: 8/28/20 12:00AM

Date Published: 11/1/19 5:00AM

Publication Location:

Article Title: Hybrid neural network potential for multilayer graphene

Authors: Mingjian Wen, Ellad B. Tadmor

Keywords: Hybrid neural network potential

Abstract: Monolayer and multilayer graphene are promising materials for applications such as electronic devices, sensors, energy generation and storage, and medicine. In order to perform large-scale atomistic simulations of the mechanical and thermal behavior of graphene-based devices, accurate interatomic potentials are required. Here, we present an interatomic potential for multilayer graphene structures referred to as “hNN-Grx.” This hybrid potential employs a neural network to describe short-range interactions and a theoretically motivated analytical term to model long-range dispersion. The potential is trained against a large dataset of monolayer graphene, bilayer graphene, and graphite configurations obtained from ab initio total-energy calculations based on density functional theory (DFT).

Distribution Statement: 2-Distribution Limited to U.S. Government agencies only; report contains proprietary info
Acknowledged Federal Support: Y

Publication Type: Journal Article

Peer Reviewed: Y

Publication Status: 1-Published

Journal: Nature

Publication Identifier Type: DOI

Publication Identifier: 10.1038/s41586-020-2458-7

Volume: 583 Issue: 7815

First Page #: 221

Date Submitted: 8/28/20 12:00AM

Date Published: 7/1/20 5:00AM

Publication Location:

Article Title: Tunable spin-polarized correlated states in twisted double bilayer graphene

Authors: Xiaomeng Liu, Zeyu Hao, Eslam Khalaf, Jong Yeon Lee, Yuval Ronen, Hyobin Yoo, Danial Haei Najafat

Keywords: spin-polarized correlated states, twisted double bilayer graphene

Abstract: Reducing the energy bandwidth of electrons in a lattice below the long-range Coulomb interaction energy promotes correlation effects. Moiré superlattices— which are created by stacking van der Waals heterostructures with a controlled twist angle^{1–3}—enable the engineering of electron band structure. Exotic quantum phases can emerge in an engineered moiré flat band. The recent discovery of correlated insulator states, superconductivity and the quantum anomalous Hall effect in the flat band of magic-angle twisted bilayer graphene^{4–8} has sparked the exploration of correlated electron states in other moiré systems^{9–11}. The electronic properties of van der Waals moiré superlattices can further be tuned by adjusting the interlayer coupling⁶ or the band structure of constituent layers⁹. Here, using van der Waals heterostructures of twisted double bilayer graphene (TDBG), we demonstrate a flat electron band that is tunable by perpendicular electric fields in a range of twist angles.

Distribution Statement: 2-Distribution Limited to U.S. Government agencies only; report contains proprietary info
Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: Physical Review Research
Publication Identifier Type: DOI **Publication Identifier:** 10.1103/PhysRevResearch.2.033162
Volume: 2 **Issue:** 3 **First Page #:**
Date Submitted: 8/15/21 12:00AM **Date Published:** 7/1/20 5:00AM
Publication Location:

Article Title: Duality between atomic configurations and Bloch states in twistrionic materials

Authors: Stephen Carr, Daniel Massatt, Mitchell Luskin, Efthimios Kaxiras

Keywords: duality, bloch states

Abstract: The relative orientation (twist) of successive layers of stacked two-dimensional (2D) materials creates variations in the interlayer atomic registry. The variations often form a superlattice, called a moiré pattern, which can alter electronic properties. In this work we introduce a classification of the single-particle electronic structures that can occur in twisted stacks of 2D layers by characterizing them as “moiré molecules” or “moiré crystals.” The molecules generate localized electronic states and moiré flat bands, while the crystals are sometimes unconventional and produce electronic banding in the configuration basis. The underpinning of this classification is the duality between interlayer configuration and monolayer Bloch momentum in moiré Hamiltonians. We apply this understanding to diagrams of local electron density in untwisted geometries to produce intuitive and quantitative predictions of twistrionic properties.

Distribution Statement: 2-Distribution Limited to U.S. Government agencies only; report contains proprietary info
Acknowledged Federal Support: Y

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: Physical Review B
Publication Identifier Type: DOI **Publication Identifier:** 10.1103/PhysRevB.102.075308
Volume: 102 **Issue:** 7 **First Page #:**
Date Submitted: 8/15/21 12:00AM **Date Published:** 8/1/20 5:00AM
Publication Location:

Article Title: Finite-size effects in wave transmission through plasmonic crystals: A tale of two scales

Authors: Matthias Maier, Mitchell Luskin, Dionisios Margetis

Keywords: wave transmission, plasmonic crystals

Abstract: The relative orientation (twist) of successive layers of stacked two-dimensional (2D) materials creates variations in the interlayer atomic registry. The variations often form a superlattice, called a moiré pattern, which can alter electronic properties. In this work we introduce a classification of the single-particle electronic structures that can occur in twisted stacks of 2D layers by characterizing them as “moiré molecules” or “moiré crystals.” The molecules generate localized electronic states and moiré flat bands, while the crystals are sometimes unconventional and produce electronic banding in the configuration basis. The underpinning of this classification is the duality between interlayer configuration and monolayer Bloch momentum in moiré Hamiltonians. We apply this understanding to diagrams of local electron density in untwisted geometries to produce intuitive and quantitative predictions of twistrionic properties.

Distribution Statement: 2-Distribution Limited to U.S. Government agencies only; report contains proprietary info
Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: Physical Review Letters
Publication Identifier Type: DOI **Publication Identifier:** 10.1103/PhysRevLett.125.116404
Volume: 125 **Issue:** 11 **First Page #:**
Date Submitted: 8/15/21 12:00AM **Date Published:** 9/1/20 5:00AM
Publication Location:

Article Title: Twisted Trilayer Graphene: A Precisely Tunable Platform for Correlated Electrons

Authors: Ziyang Zhu, Stephen Carr, Daniel Massatt, Mitchell Luskin, Efthimios Kaxiras

Keywords: twisted, trilayer, graphene

Abstract: The relative orientation (twist) of successive layers of stacked two-dimensional (2D) materials creates variations in the interlayer atomic registry. The variations often form a superlattice, called a moiré pattern, which can alter electronic properties. In this work we introduce a classification of the single-particle electronic structures that can occur in twisted stacks of 2D layers by characterizing them as “moiré molecules” or “moiré crystals.” The molecules generate localized electronic states and moiré flat bands, while the crystals are sometimes unconventional and produce electronic banding in the configuration basis. The underpinning of this classification is the duality between interlayer configuration and monolayer Bloch momentum in moiré Hamiltonians. We apply this understanding to diagrams of local electron density in untwisted geometries to produce intuitive and quantitative predictions of twistrionic properties.

Distribution Statement: 2-Distribution Limited to U.S. Government agencies only; report contains proprietary info
Acknowledged Federal Support: Y

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: Journal of Computational Physics
Publication Identifier Type: DOI **Publication Identifier:** 10.1016/j.jcp.2020.109871
Volume: 423 **Issue:** **First Page #:** 109871
Date Submitted: 8/15/21 12:00AM **Date Published:** 12/1/20 6:00AM
Publication Location:

Article Title: Nonlinear eigenvalue problems for coupled Helmholtz equations modeling gradient-index graphene waveguides

Authors: Jung Heon Song, Matthias Maier, Mitchell Luskin

Keywords: Guided mode Time-harmonic Maxwell's equations Surface plasmon polariton

Abstract: The relative orientation (twist) of successive layers of stacked two-dimensional (2D) materials creates variations in the interlayer atomic registry. The variations often form a superlattice, called a moiré pattern, which can alter electronic properties. In this work we introduce a classification of the single-particle electronic structures that can occur in twisted stacks of 2D layers by characterizing them as “moiré molecules” or “moiré crystals.” The molecules generate localized electronic states and moiré flat bands, while the crystals are sometimes unconventional and produce electronic banding in the configuration basis. The underpinning of this classification is the duality between interlayer configuration and monolayer Bloch momentum in moiré Hamiltonians. We apply this understanding to diagrams of local electron density in untwisted geometries to produce intuitive and quantitative predictions of twistrionic properties.

Distribution Statement: 2-Distribution Limited to U.S. Government agencies only; report contains proprietary info
Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: Physical Review B
Publication Identifier Type: DOI **Publication Identifier:** 10.1103/PhysRevB.102.205411
Volume: 102 **Issue:** 20 **First Page #:**
Date Submitted: 8/15/21 12:00AM **Date Published:** 11/1/20 5:00AM
Publication Location:

Article Title: Dipole excitation of collective modes in viscous two-dimensional electron systems

Authors: Vera Andreeva, Denis A. Bandurin, Mitchell Luskin, Dionisios Margetis

Keywords: Dipole excitation, 2D

Abstract: The relative orientation (twist) of successive layers of stacked two-dimensional (2D) materials creates variations in the interlayer atomic registry. The variations often form a superlattice, called a moiré pattern, which can alter electronic properties. In this work we introduce a classification of the single-particle electronic structures that can occur in twisted stacks of 2D layers by characterizing them as “moiré molecules” or “moiré crystals.” The molecules generate localized electronic states and moiré flat bands, while the crystals are sometimes unconventional and produce electronic banding in the configuration basis. The underpinning of this classification is the duality between interlayer configuration and monolayer Bloch momentum in moiré Hamiltonians. We apply this understanding to diagrams of local electron density in untwisted geometries to produce intuitive and quantitative predictions of twistrionic properties.

Distribution Statement: 2-Distribution Limited to U.S. Government agencies only; report contains proprietary info
Acknowledged Federal Support: Y

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: Multiscale Modeling & Simulation
Publication Identifier Type: DOI **Publication Identifier:** 10.1137/19M1273499
Volume: 18 **Issue:** 4 **First Page #:** 1525
Date Submitted: 8/15/21 12:00AM **Date Published:** 12/1/20 6:00AM
Publication Location:

Article Title: Modeling and Computation of Kubo Conductivity for Two-Dimensional Incommensurate Bilayers

Authors: Simon Etter, Daniel Massatt, Mitchell Luskin, Christoph Ortner

Keywords: two-dimensional materials, Kubo, Chebyshev

Abstract: This paper presents a unified approach to the modeling and computation of the Kubo conductivity of incommensurate bilayer heterostructures at finite temperature. First, we derive an expression for the large-body limit of Kubo--Greenwood conductivity in terms of an integral of the conductivity function with respect to a current-current correlation measure. We then observe that the incommensurate structure can be exploited to decompose the current-current correlation measure into local contributions and deduce an approximation scheme which is exponentially convergent in terms of domain size. Second, we analyze the cost of computing local conductivities via Chebyshev approximation. Our main finding is that if the inverse temperature β is sufficiently small compared to the inverse relaxation time η , namely $\beta \lesssim \eta^{-1/2}$, then the dominant computational cost is $\mathcal{O}(\eta^{-3/2})$ inner products for a suitably truncated Chebyshev series, which significantly improves on t

Distribution Statement: 2-Distribution Limited to U.S. Government agencies only; report contains proprietary info
Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Journal of Mathematical Physics

Publication Identifier Type: DOI

Publication Identifier: 10.1063/5.0021961

Volume: 62

Issue: 1

First Page #: 011503

Date Submitted: 8/15/21 12:00AM

Date Published: 1/1/21 6:00AM

Publication Location:

Article Title: Homogenization of hydrodynamic transport in Dirac fluids

Authors: Guillaume Bal, Andrew Lucas, Mitchell Luskin

Keywords: homogenization, hydrodynamic transport

Abstract: Large-scale electrical and thermal currents in ordinary metals are well approximated by effective medium theory: global transport properties are governed by the solution to homogenized coupled diffusion equations. In some metals, including the Dirac fluid of nearly charge neutral graphene, microscopic transport is not governed by diffusion, but by a more complicated set of linearized hydrodynamic equations, which form a system of degenerate elliptic equations coupled with the Stokes equation for fluid velocity. In sufficiently inhomogeneous media, these hydrodynamic equations reduce to homogenized diffusion equations. We recast the hydrodynamic transport equations as the infimum of a functional over conserved currents and present a functional framework to model and compute the homogenized diffusion tensor relating electrical and thermal currents to charge and temperature gradients. We generalize to this system to the well-known results in homogenization theory: Tartar's proof of local

Distribution Statement: 2-Distribution Limited to U.S. Government agencies only; report contains proprietary info
Acknowledged Federal Support: Y

Publication Type: Journal Article

Peer Reviewed: Y

Publication Status: 1-Published

Journal: Science

Publication Identifier Type: DOI

Publication Identifier: 10.1126/science.abg0399

Volume: 371

Issue: 6534

First Page #: 1133

Date Submitted: 8/15/21 12:00AM

Date Published: 3/1/21 6:00AM

Publication Location:

Article Title: Electric field–tunable superconductivity in alternating-twist magic-angle trilayer graphene

Authors: Zeyu Hao, A. M. Zimmerman, Patrick Ledwith, Eslam Khalaf, Danial Haie Najafabadi, Kenji Watanabe, T

Keywords: superconductivity, trilayer graphene

Abstract: The discovery of superconductivity in bilayers consisting of graphene sheets twisted with respect to each other by just the right “magic” angle has inspired enormous interest in twisted materials. Hao et al. constructed twisted trilayer graphene, in which the middle layer is twisted with respect to the bottom layer and the top layer is roughly parallel to the bottom layer (see the Perspective by Yazdani). Such trilayers exhibited superconductivity that was tunable by an external electric field and consistent with having an unconventional nature.

Distribution Statement: 2-Distribution Limited to U.S. Government agencies only; report contains proprietary info
Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Physical Review B

Publication Identifier Type: DOI

Publication Identifier: 10.1103/PhysRevB.102.125424

Volume: 102

Issue: 12

First Page #:

Date Submitted: 8/18/21 12:00AM

Date Published: 9/1/20 5:00AM

Publication Location:

Article Title: Lithium intercalation in MoS₂ bilayers and implications for moiré flat bands

Authors: Zheyu Lu, Stephen Carr, Daniel T. Larson, Efthimios Kaxiras

Keywords: Lithium intercalation

Abstract: Intercalation of lithium atoms between layers of two-dimensional (2D) materials can alter their atomic and electronic structure. We investigate effects of Li intercalation in twisted bilayers of the transition metal dichalcogenide MoS₂ through first-principles calculations, tight-binding parametrization based on the Wannier transformation, and analysis of moiré band structures through an effective continuum model. The energetic stability of different intercalation sites for Li between layers of MoS₂ are classified according to the local coordination type and the number of vertically aligned Mo atoms, suggesting that the Li atoms will cluster in certain regions of the moiré superlattice. The proximity of a Li atom has a dramatic influence on the interlayer interaction between sulfur atoms, deepening the moiré potential well and leading to better isolation of the flat bands in the energy spectrum.

Distribution Statement: 2-Distribution Limited to U.S. Government agencies only; report contains proprietary info

Acknowledged Federal Support: Y

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Nature Communications

Publication Identifier Type: DOI

Publication Identifier: 10.1038/s41467-020-20428-1

Volume: 12

Issue: 1

First Page #:

Date Submitted: 8/18/21 12:00AM

Date Published: 1/1/21 6:00AM

Publication Location:

Article Title: Moiré metrology of energy landscapes in van der Waals heterostructures

Authors: Dorri Halbertal, Nathan R. Finney, Sai S. Sunku, Alexander Kerelsky, Carmen Rubio-Verdú, Sara Shab

Keywords: Moiré metrology

Abstract: The emerging field of twistronics, which harnesses the twist angle between two-dimensional materials, represents a promising route for the design of quantum materials, as the twistangle- induced superlattices offer means to control topology and strong correlations. At the small twist limit, and particularly under strain, as atomic relaxation prevails, the emergent moiré superlattice encodes elusive insights into the local interlayer interaction. Here we introduce moiré metrology as a combined experiment-theory framework to probe the stacking energy landscape of bilayer structures at the 0.1 meV/atom scale, outperforming the gold-standard of quantum chemistry. Through studying the shapes of moiré domains with numerous nano-imaging techniques, and correlating with multi-scale modelling, we assess and refine first-principle models for the interlayer interaction.

Distribution Statement: 2-Distribution Limited to U.S. Government agencies only; report contains proprietary info

Acknowledged Federal Support: Y

RPPR Final Report as of 16-Mar-2022

Publication Type: Journal Article

Peer Reviewed: Y

Publication Status: 1-Published

Journal: Physical Review Letters

Publication Identifier Type: DOI

Publication Identifier: 10.1103/PhysRevLett.127.166802

Volume: 127 Issue: 16

First Page #:

Date Submitted: 3/5/22 12:00AM

Date Published: 10/1/21 5:00AM

Publication Location:

Article Title: Correlated Insulating States and Transport Signature of Superconductivity in Twisted Trilayer Graphene Superlattices

Authors: Xi Zhang, Kan-Ting Tsai, Ziyang Zhu, Wei Ren, Yujie Luo, Stephen Carr, Mitchell Luskin, Efthimios Kaxiras

Keywords: Correlating, Insulating, Superconductivity

Abstract: Layers of two-dimensional materials stacked with a small twist angle give rise to beating periodic patterns on a scale much larger than the original lattice, referred to as a “moiré superlattice.” Here, we demonstrate a higher-order “moiré of moiré” superlattice in twisted trilayer graphene with two consecutive small twist angles. We report correlated insulating states near the half filling of the moiré of moiré superlattice at an extremely low carrier density.

Distribution Statement: 1-Approved for public release; distribution is unlimited.

Acknowledged Federal Support: Y

Publication Type: Journal Article

Peer Reviewed: Y

Publication Status: 1-Published

Journal: Journal of Mathematical Physics

Publication Identifier Type: DOI

Publication Identifier: 10.1063/5.0054122

Volume: 62 Issue: 9

First Page #: 091502

Date Submitted: 3/5/22 12:00AM

Date Published: 9/1/21 5:00AM

Publication Location:

Article Title: Existence of the first magic angle for the chiral model of bilayer graphene

Authors: Alexander B. Watson, Mitchell Luskin

Keywords: magic angle, graphene, twisted

Abstract: Layers of two-dimensional materials stacked with a small twist angle give rise to beating periodic patterns on a scale much larger than the original lattice, referred to as a “moiré superlattice.” Here, we demonstrate a higher-order “moiré of moiré” superlattice in twisted trilayer graphene with two consecutive small twist angles. We report correlated insulating states near the half filling of the moiré of moiré superlattice at an extremely low carrier density.

Distribution Statement: 1-Approved for public release; distribution is unlimited.

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I certify that the information in the report is complete and accurate:

Signature: Mitchell Luskin

Signature Date: 3/15/22 11:25AM

Accomplishments

Relaxation and domain formation model for incommensurate two-dimensional heterostructures

In [5], we introduced configuration space as a natural representation for calculating the mechanical relaxation patterns of incommensurate two-dimensional (2D) bilayers. Our approach has now been applied to a wide variety of 2D twisted bilayer materials through the use of a continuum model in combination with a generalized stacking fault energy for interlayer interactions. We calculated accurate relaxations for graphene and MoS₂ heterostructures even at small twist-angle values, enabled by the fact that our approach does not rely on empirical atomistic potentials for interlayer coupling.

The advantages of our approach are twofold: first, to study continuously and rigorously the limit of small angles where large-scale moiré structures appear; second, to avoid the massive computational cost associated with atomic simulations in supercells in the corresponding periodic approximations for which the area (and number of atoms) scales as the squared inverse of the twist angle. The almost periodic patterns being computed from this new theory is the input needed to compute the density of states and transport properties using the recently developed electronic structure theory and numerical methods of incommensurate 2D materials.

We further gave a general theory for the relaxation of multilayer incommensurate heterostructures in [8] that can be applied to any number of layers. We applied this theory to incommensurate trilayer van der Waals heterostructures in [10] and used it to explore the “moiré of moiré” relaxation patterns of twisted trilayer graphene and WSe₂.

Concurrently, we carried out an extensive experimental investigation of atomic and electronic reconstruction at the van der Waals interface in twisted bilayer graphene in [6] and compared the experimental results with those obtained by our theoretical model.

We have expanded our multi-scale theoretical modeling of twisted multi-layered materials. In terms of theoretical tools, we have derived a minimal symmetry-preserving model for twisted bilayer graphene. The minimal model makes it possible to estimate electron interaction strength and construct a self-consistent model that includes electronic interaction at the moiré scale. We also studied more types of twisted multi-layered materials, beyond twisted bilayer graphene. The twisted sandwich graphene with a twisted middle layer exhibits a Dirac cone and flat bands near the charge-neutrality in proximity, which we have shown to be experimentally realizable and to have the potential to host more exotic strongly correlated states.

We also systematically studied a fundamentally different type of system – twisted trilayers with two-independent twist angles. The system exhibits higher-order moiré of moiré patterns and they are incommensurate even in the continuum limit. The long length scale and the aperiodicity make it difficult to characterize the system based on a real space model. For the mechanical relaxation, we developed an energy minimization model in configuration space based on first principles calculations. Configuration space describes the local environment of atomic positions, which is a four-dimensional space in moiré of moiré twisted trilayers. This approach bypasses the need for a periodic supercell. We show that the relaxation pattern in twisted trilayers can break the out-of-plane mirror symmetry. For the electronic

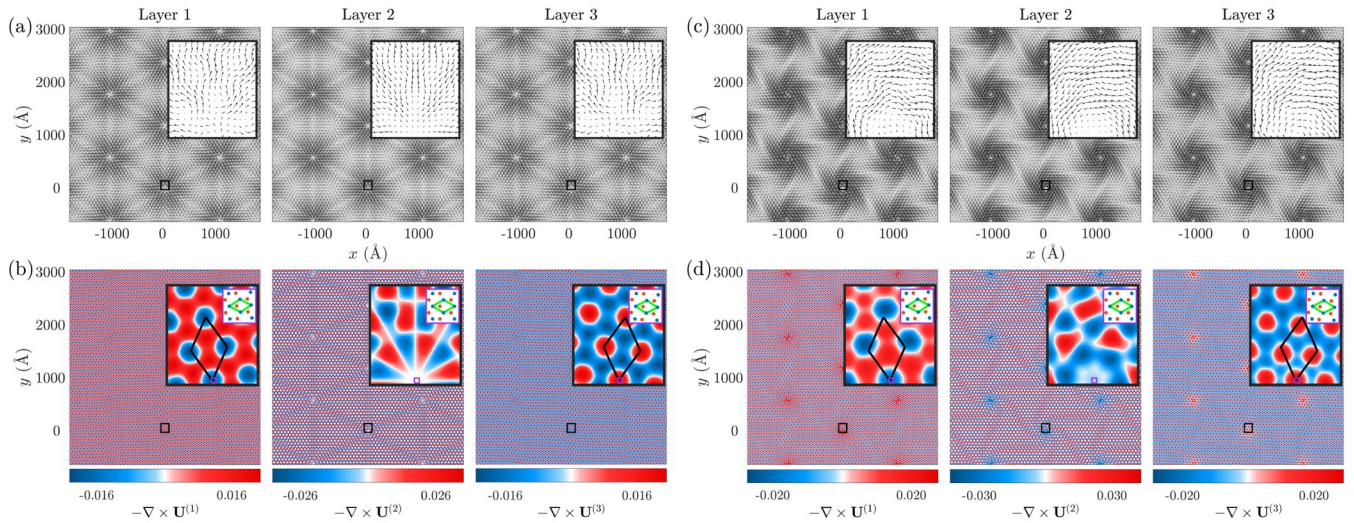


Figure 1 Relaxation pattern of twisted trilayer WSe2 with $\theta_{12} = \theta_{23} = 3^\circ$. Top: the relaxation displacement vectors; bottom: the curl of displacements. Insets are the blow up of the black boxed regions. (a)-(b) shows the symmetric solution and (c)-(d) shows the asymmetric solution.

structure model, we employed a low-energy momentum space model for twisted trilayer graphene to study the evolution of van Hove singularities. We show that the system exhibits a wide range magic angles when twist angles are different and exists a regime where the two bilayer moiré supercells strongly hybridize (Fig. 2). Our results show that the additional tuning “knob” – a second twist angle – leads to much richer electronic behaviors.

Commensuration and Incommensuration in van der Waals Heterojunctions

Heterogeneous interfaces between two dissimilar materials are an essential building block for modern semiconductor devices. The 2-dimensional (2D) van der Waals (vdW) materials and their heterostructures provide a new opportunity to realize atomically sharp interfaces in the ultimate quantum limit for the electronic and optoelectronic processes. By assembling atomic layers of vdW materials, such as hexa boronitride, transition metal chalcogenide and graphene, we can construct atomically thin novel quantum structures. We demonstrate the enhanced electronic and optoelectronic performances in the vdW heterostructures, suggesting that these a few atom thick interfaces may provide a fundamental platform to realize novel physical phenomena. In this presentation, we will discuss transmission electron microscopy (TEM) investigation of the microstructures of 2-D based

heterostructures. Microstructural properties of the heterostructures caused by the interlayer interaction were studied by TEM-based analytical techniques such as electron diffraction, dark field imaging, and aberration corrected scanning TEM imaging. We show that the commensurate domain structures can be formed as a result of the interplay between van der Waals interaction energy and elastic energy of individual layers. Based on the experimental data, we identify microstructural origin that can cause the structural transition in the system.

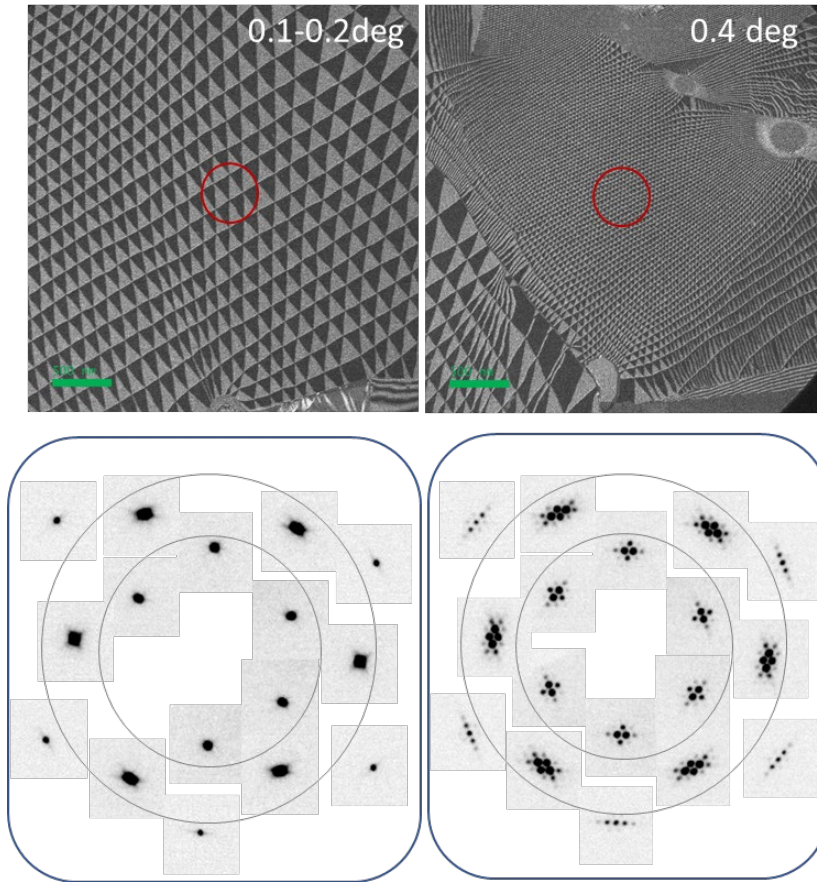


Fig. P2: Upper panels show the dark field TEM image showing the reconstructed Moiré patterns in twisted graphene bilayers. Bottom panel shows the corresponding selected area diffraction pattern that shows the microscopic details of atomic reconstruction.

Real space electronic structure model for incommensurate 2D materials

We utilized our configuration space approach to develop the first real space models to compute the density of states and transport properties for incommensurate 2D multilayer materials without periodic or continuum approximations. Our real space model gives the local density of states associated with each configuration and then integrates over the set of possible configurations to give the density of states [1,2]. We developed an efficient solver for the local density of states that utilizes the kernel approximation method to avoid the costly diagonalization of large Hamiltonians.

We extended our real space Kubo approach to the modeling and computation of the Kubo conductivity of incommensurate bilayer heterostructures at finite temperature. We derived an expression for the large-body limit of Kubo--Greenwood conductivity in terms of an integral of the conductivity function with respect to a current-current correlation measure [3,12]. We then observed that the incommensurate structure can be exploited to decompose the current-current correlation measure into local contributions and deduced an approximation scheme which is exponentially convergent in terms of domain size [12].

Momentum space electronic structure model for incommensurate 2D materials

We have developed a momentum space model for the electronic structure and transport properties of incommensurate 2D materials that is the dual of the real space model [4]. We formulated a Hamiltonian on the incommensurate reciprocal lattice that is dual to the Hamiltonian of the real space lattice. The momentum space approach usually gives a more efficient computational method since the local density of states in momentum space is concentrated in a small region of momentum space (near the Dirac cones in graphene) that leads to much smaller matrices to diagonalize and to a smaller set of momentum to sample to compute the density of states [4] and conductivity [9].

Rigorous proof of the existence of flat bands for the chiral model.

Motivated by recent experimental discoveries of correlated superconducting phases for twisted bilayer graphene at “magic angles” that give nearly flat bands for the Bistritzer-MacDonald model, we gave a rigorous proof of the existence of a zero Fermi velocity at the moiré K point at the first magic angle for the chiral model [14]. Our results combined with the proof of Tarnopolsky-Kruchkov-Vishwanath of the existence of a perfectly flat band at zero energy over the whole Brillouin zone for twist angles for which the effective Fermi velocity at the moiré K point vanishes, proves that the chiral model has a perfectly flat band.

Development of measuring electronic thermal transport in graphene/hBN heterostructures.

Interactions between particles in quantum many-body systems can lead to collective behavior described by hydrodynamics. One such system is the electron-hole plasma in graphene near the charge neutrality point which can form a strongly coupled Dirac fluid. This charge neutral plasma of quasi-relativistic fermions is expected to exhibit a substantial enhancement of the thermal conductivity, due to decoupling of charge and heat currents within hydrodynamics. Employing sensitive Johnson noise thermometry, we report the breakdown of the Wiedemann-Franz law in graphene, with a thermal conductivity an order of magnitude larger than the value predicted by Fermi liquid theory. This result is a signature of the Dirac fluid and constitutes direct evidence of collective motion in a quantum electronic fluid.

Homogenization of hydrodynamic transport in Dirac fluids and the breakdown of the Wiedemann-Franz law

Motivated by recent experimental results of the Kim Group on the breakdown in clean graphene of the Wiedemann-Franz law that the electronic contribution of the thermal conductivity is proportional to the temperature [15], we have given a rigorous proof that in a Dirac fluid of nearly charge neutral graphene, microscopic transport is not governed by diffusion, but by a more complicated set of linearized hydrodynamic equations, which form a system of degenerate elliptic equations coupled with

the Stokes equation for fluid velocity [13]. In sufficiently inhomogeneous media, these hydrodynamic equations reduce to homogenized diffusion equations. We recast the hydrodynamic transport equations as the infimum of a functional over conserved currents and presented a functional framework to model and compute the homogenized diffusion tensor relating electrical and thermal currents to charge and temperature gradients.

Modeling for Strained TMDCs

The layered materials provide a nice platform to study the interplay between different physics and potential device applications. One of the control knobs to tune the desired material properties is the strain engineering, which can cause the band gaps, bandwidth and other electronic or optical properties to change and also lower the crystal symmetry. Recently, the anisotropic strain field is also applied to study the many-body correlated phase such as fractional quantum Hall liquid. TMDCs are a class of interesting materials with applications in optoelectronics, valleytronics and spintronics. In our work, we study the modeling of strained hamiltonian in these TMDCs layers. By the group representation analysis, we identify the strain coupling terms and parametrize their coupling strength based on Wannier analysis of density functional theory calculations. We further derive the low-energy kp hamiltonians and identify the pseudogauge field terms. Besides the above H structure type of TMDCs, we also generalize to the T structure TMDCs. These T-type TMDCs can host different physics properties (such as superconductivity and charge density wave in NbSe₂ and TaS₂). Recently, the quantum spin liquid phase is speculated in the T-TaS₂ in the low-temperature CDW and Mott phase. Stephen is also working on integrating the structural relaxation with the strained tight-binding model derived above, which is important for moire systems such as twisted bilayer graphene with small rotation angle.

Superlattice-induced insulating states and valley-protected orbits in twisted bilayer graphene:

Transport measurement of twisted bilayer graphene (~ 1.8 degree) done in MIT Jarillo-Herrero's group shows (surprisingly) insulating behavior at certain moire superlattice filling which can be attributed to the single particle gap, which is in agreement with our ab-initio Wannier tight-binding modeling. This strongly suggests the importance of interlayer coupling in the low-energy theory of vdW heterostructures. Further progress is on the sample with even smaller twist angle, such as 1.1 degree at magic angle where flat bands are expected and ~ 0.6 degree close to second magic angle where another set of flat bands are close. We are investigating a range of issues such as single particle electronic structure, interaction effects from moire spots, and hofstadter butterfly fractal effects in magneto-transport. The kernel polynomial method developed for the incommensurate systems will be crucial for us to understand these twisted systems and their interactions to optical excitations and the magnetic field.

Wannier function decompositions

In our previous works, we have demonstrated the reliable tight-binding modeling constructions based on the Wannier function transformation of density functional theory calculations. We applied this method to the layered materials and model the intralayer/interlayer couplings, and their response to the strain field. The next challenge is how to apply the technique to study the interlayer properties in between different kinds of layered materials. One way is to extract the Wannier orbitals from each layers and compute the relevant overlap integrals. This work is a collaboration with the Luskin and Cancès groups, who are developing efficient Wannier function decomposition methods. Their numerical

technique is accurate, and suitable to be applied later to model the interlayer coupling by evaluating relevant hamiltonian matrix elements.

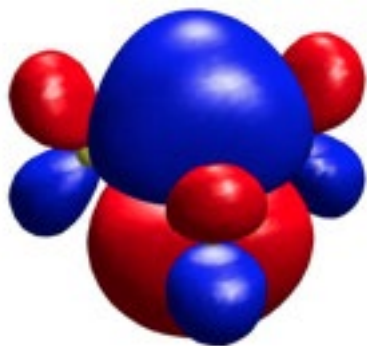


Figure 2. Graphene p_z Wannier orbital, with the three-fold structure from crystal field distortion.

Modeling of in-plane graphene/MoS2 heterojunctions:

In our previous work published in Nano Letters, we collaborated with Prof. T. Palacios's group at MIT. A MoS₂ field-effect transistor with sub-10 nm channel length was successfully fabricated and exhibited remarkable performance. We used density functional theory calculations to study the stability of the semiconducting 2H and metallic 1T' phases, and the electronic properties at the boundary formed by joining the two phases laterally. In this project, we study the graphene/MoS₂ lateral heterojunction, which promises to exhibit a low-resistance contact according to the Schottky-Mott rule. We provided an atomic-scale description of the structural, electronic, and magnetic properties of this type of junction. We identified the energetically favorable structures in which the preference of forming C-S or C-Mo bonds at the boundary depends on the chemical conditions. We found that significant charge transfer between graphene and MoS₂ is localized at the boundary. We also showed that the abundant 1D boundary states substantially pin the Fermi level in the lateral contact between graphene and MoS₂, in close analogy to the effect of 2D interfacial states in the contacts between 3D materials. Furthermore, we proposed specific ways in which these effects can be exploited to achieve spin-polarized currents. We are currently investigating the topological properties of the MoS₂ edges and the graphene/MoS₂ boundaries.

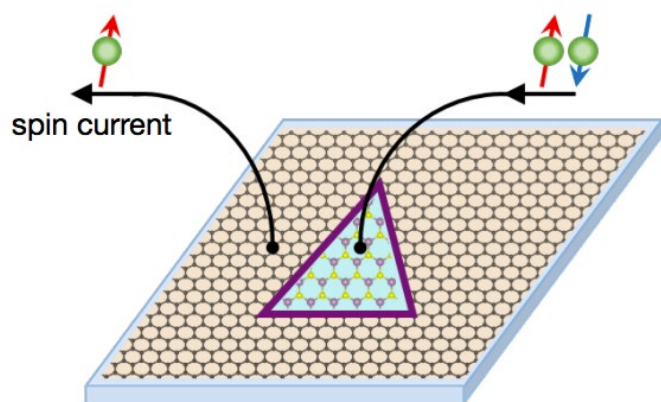


Figure 3: A schematic of spin-polarized current generation using graphene/MoS2 in-plane heterojunctions.

Engineering quantum confinement of charge carriers and charged exciton in 2D semiconductors:

Electrical confinement and manipulation of charge carriers in semiconducting nanostructures are essential for realizing functional quantum electronic devices. The unique band structure of atomically thin transition metal dichalcogenides (TMDs), offers a new route towards realizing novel 2-dimensional (2D) quantum electronic devices, such as valleytronic devices and valley-spin qubits. 2D TMDs also provide a platform for novel quantum optoelectronic devices due to large exciton binding energy. However, controlled confinement and manipulation of electronic and excitonic excitations in TMD nanostructures have been technically challenging due to the prevailing disorder in the material, preventing accurate experimental control of local confinement and tunnel couplings. In this work, we demonstrate a novel method for creating high-quality heterostructure composed from atomically thin materials that allows for efficient electrical control of excitations. Specifically, we demonstrate quantum transport in the gate-defined quantum-confined region, observing spin-valley locked quantized conductance in quantum point contacts (QPCs). We also realize gate-controlled Coulomb blockade associated with confinement of electrons and demonstrate electrical control over charged excitons with tunable local confinement potentials and tunnel couplings. Our work provides a basis for novel quantum opto-electronic devices based on manipulation of electrons and charged excitons.

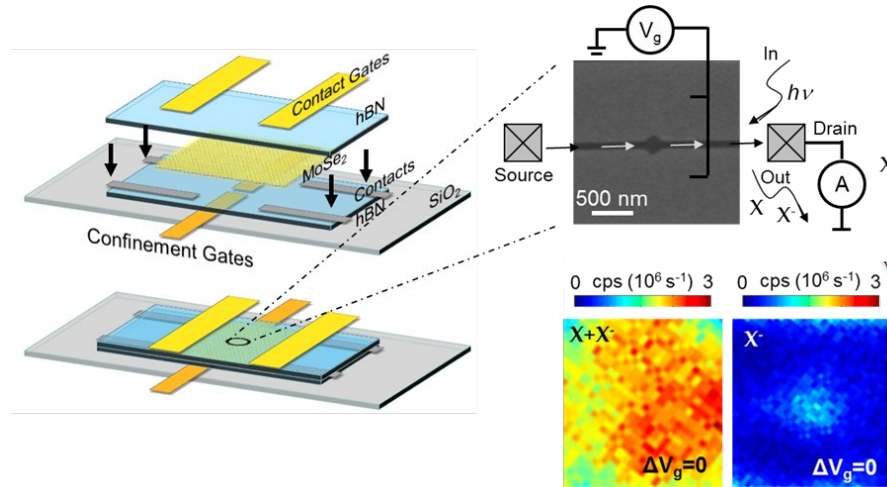


Fig. P1: Left panel shows a schematic diagram of device built for confinement of charged carriers and excitons using electric gate. The right panel shows the wiring diagram for gate control and spatial maps of the optical spectrum of excitons and trions.

Epsilon-Near-Zero in plasmonic crystal composed by 2D conductors and spatial dependent dielectric host

We calculate analytically the isofrequency k -space band structure of a plasmonic crystal consists of 2D conductors, like doped graphene, embedded in a periodic array of a dielectric with space depended dielectric constant. We find that a Plasmonic Dirac point always exists leading to Epsilon-Near-Zero

behavior. Moreover, we calculate exactly the critical ENZ period in terms of the dielectric function characteristics

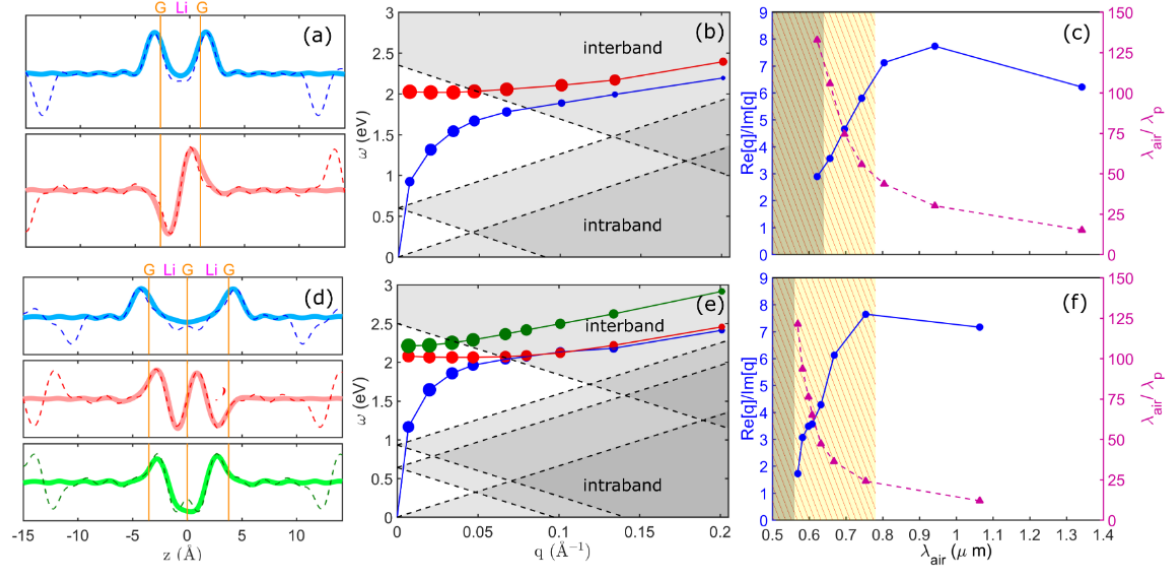


Figure 11. Images (a) and (d) indicate the plasmon modes in bilayer and trilayer, respectively. Images (b) and (e) show the plasmon dispersion relation in bi- and tri-layer and (c) and (f) present the plasmon localization and propagation length in bilayer and trilayer, respectively.

Visible quantum plasmons in in highly-doped few-layer graphene:

We investigated the existence of visible plasmons in highly doped bilayer and trilayer of graphene. We show, through the use of first-principles calculations, that the high levels of doping achieved by lithium intercalation in bilayer and trilayer graphene shift the plasmon frequencies into the visible range. We describe the features of these quantum plasmons (Fig. 11 a, d), including the dispersion relation (Fig. 11 b, e), losses and field localization (Fig. 11 c, f). Our findings point to a strategy for fine-tuning the plasmon frequencies in graphene and other two dimensional materials. In addition, our results expand the frequency range of the Dirac plasmons, which was restricted up to mid-infrared regime. A manuscript including all the above theoretical finding has been submitted for publication.

Properties of In-Plane Graphene/MoS2 Heterojunctions

In this project, we study the graphene/MoS2 lateral heterojunction, which promises to exhibit a low-resistance contact according to the Schottky-Mott rule. We provided an atomic-scale description of the structural, electronic, and magnetic properties of this type of junction. We identified the energetically favorable structures in which the preference of forming C-S or C-Mo bonds at the boundary depends on the chemical conditions. We found that significant charge transfer between graphene and MoS2 is localized at the boundary. We also showed that the abundant 1D boundary states substantially pin the Fermi level in the lateral contact between graphene and MoS2, in close analogy to the effect of 2D interfacial states in the contacts between 3D materials. Furthermore, we proposed specific ways in

which these effects can be exploited to achieve spin-polarized currents. We are currently investigating the topological properties of the MoS₂ edges and the graphene/MoS₂ boundaries.

Effects of magnetic dopants in (Li_{0.8}M_{0.2}OH)FeSe (M = Fe, Mn, Co)

In this project, we collaborated with Prof. Z. Zhang from University of Science and Technology of China and Prof. M. Weinert from University of Wisconsin. We investigated the effects of Fe dopants on the electronic bands structure of (Li_{0.8}Fe_{0.2}OH)FeSe using a band unfolding technique and first-principles supercell calculations. The band structure for the surface FeSe layer with checkerboard antiferromagnetic order is reasonably consistent with angle-resolved photoemission results. The 3d transition metals Mn and Co have similar doping effects on the band structure of (LiOH)FeSe.

Twisted bilayer graphene: experimental collaborations

The moiré superlattice in two-dimensional layered materials can greatly modify the electronic properties of the system. Previously we studied the twisted bilayer graphene system at larger twist angle (about 2 degree). The alternative interference pattern of the twisted crystal mostly renormalize the monolayer Dirac electrons and reduce the Fermi velocity. The interlayer coupling potential also generates additional insulating states when electron or hole doped away from charge neutrality (Y. Can et al., Phys. Rev. Lett. 117, 116804 (2016)) due to the formation of single particle gaps. When the twist angle is decreased, the Fermi velocity continues to drop and at around 1.1 degree (the magic angle) the flat bands emerge which was shown from the numerical simulation. In the experiment performed in Jarillo-Herrero's group in MIT, the twisted bilayer graphene at around magic angles shows Mott-like insulating states at integer fillings of moiré superlattice away from charge neutrality and superconductivity (in close proximity to the Mott-insulating states) at half-filled hole doping (Nature 556, 80 (2018) and Nature 556, 43 (2018)). The discovery might provide new insights into the high temperature superconductivity and various quantum correlated phases. Further investigations are ongoing now for the nature of the correlated insulating and the superconductivity states. One proposal we made to change the energy scale is to vary the interlayer coupling by applying pressure to the twisted bilayer (arXiv:1806.05078), which would effectively modify the magic angle and hence the characteristic density and energy scale for superconductivity. The scaling behavior can provide us with the additional tuning knob to control and probe the quantum phase. Additional experiments (STM, optical/plasmon, etc) are needed to further elucidate the issues related to the symmetry, symmetry breaking and orders in the crystal.

Advances in our ab-initio numerical

The interest to generalize the magic angle physics in graphene to other two-dimensional materials relies on the multi-scale simulations to the moiré superlattice crystal system, which has enormous number of atoms. The microscopic theory is based on ab initio numerical modeling technique combining density functional theory (DFT) calculations and Wannier transformation to derive efficient and accurate ab-initio tight-binding Hamiltonians (Phys. Rev. B 93, 235153 (2016)). The effective models can then be derived and extensive numerical simulations can be carried out with proper mathematical tool (in collaboration with my colleague Stephen Carr.) In the past, we have applied the numerical method to H-

type transition metal dichalcogenides (TMDC) and now we are working on further generalizations to other types of layers such as T-type TMDCs (in collaboration with colleague Daniel Larson) and more including the deformation of monolayer from strains (arXiv: 1709.07510), charge density waves as in many TMDC crystals (Phys. Rev. B 98, 035203 (2018)) and the interlayer couplings and their models. The accurate electronic structure modeling can be helpful in elucidating the quantum geometry properties of the electrons which are relevant to transport and optical properties of the crystal. For example, while the Berry curvature and its momentum integration is related to the Hall response of the crystal, the time-reversal invariant crystal can display non-linear Hall response due to the presence of non-zero Berry curvature dipole moment (arXiv:1805.02157).

First-principles study of vibrational and optical properties of Janus transition metal dichalcogenides

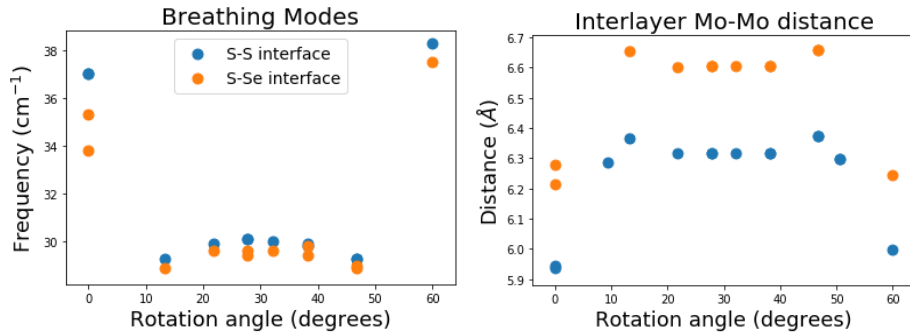


Figure 1: left: calculated layer breathing mode frequency as a function of the twist angle in Janus-MoS₂ heterostructure. **Right:** the interlayer spacing as a function of the twist angle

Using first principles density functional theory (DFT) calculations we have studied the effects of the twist angle and interface chemistry on the vibrational and optical properties of Janus transition metal dichalcogenide (TMDC) bilayers, specifically MoSSe/MoS₂. We have shown a clear inverse dependence of the interlayer breathing mode Raman frequency on the interlayer Mo-Mo distance, as shown in Fig. 1. This interlayer distance changes with the relative twist angle between the MoSSe and MoS₂ layers.

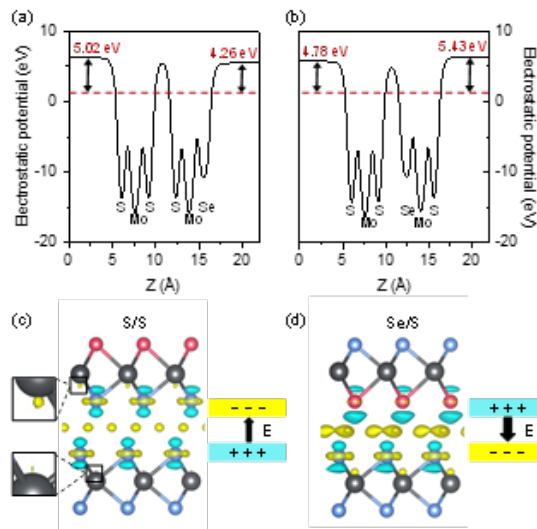


Figure 2: top: electrostatic potential for Janus-MoS₂ heterostructures with (a) S-S interface and (b) S-Se interface. **Bottom:** charge density distribution of the system with (a) S-S interface and (b) S-Se interface

An interesting feature of the Janus TMDCs like MoSSe is the charge asymmetry across the layer that leads to an intrinsic dipole moment within the layer and thus a different work function on the two sides of the bilayer heterostructure, which we were able to calculate using DFT, as shown in the top panels of Fig. 2. We also find clear differences in electronic band structure and charge density distribution depending on the species of atoms at the interface between the layers, as shown in the bottom panels.

By studying the microscopic details of the interactions between atomic species at the interface between two layers and the

dependence on the twist angle between the 2: Large data set approaches

Data-driven studies of two dimensional magnetic materials

In this project we use a data-driven approach to study the magnetic and thermodynamic properties of two-dimensional (2D) materials. Working with a collaborator, Wei Chen, we investigate over 4,000 monolayer structures based on the known ferromagnetic material Cr₂Ge₂Te₆, using density functional theory (DFT) calculations and machine learning methods (arXiv: 1806.07989). We determine their magnetic properties, such as magnetic order and magnetic moment. We also calculate the formation energies and use them as a proxy for chemical stability. We demonstrate that machine learning tools, combined with DFT calculations, can allow for the rapid prediction of properties of 2D magnetic materials. Furthermore, our data-driven approach can provide insights into the microscopic origins of magnetic ordering in these systems. Despite the possibility of the magnetic ordering of two-dimensional magnetic materials being destroyed by thermal fluctuations (Mermin-Wagner theorem), magnetocrystalline anisotropy drives the ordering of spins in reduced dimensions. Our study is a systematic investigation of 2D magnetic materials which explores new methods for the rapid discovery of chemically stable 2D materials with magnetic order.

Mathematical method and Database construction

We have been collecting the massive amount of data from simulation on atomic coupling under various crystal environment into the material tight-binding database. In the near future, we will be attempting to build numerical/statistical model for predicting atomic coupling relevant for these van der Waals heterostructure involving two-dimensional layers and their more crystals. On the other hand, developments for mathematical tools are underway to represent the Wannier functions into efficient basis for numerical purposes (Computer Physics Communications 230, 27 (2018)) and numerical schemes to compute the hopping and overlap integral involving various Wannier orbitals.

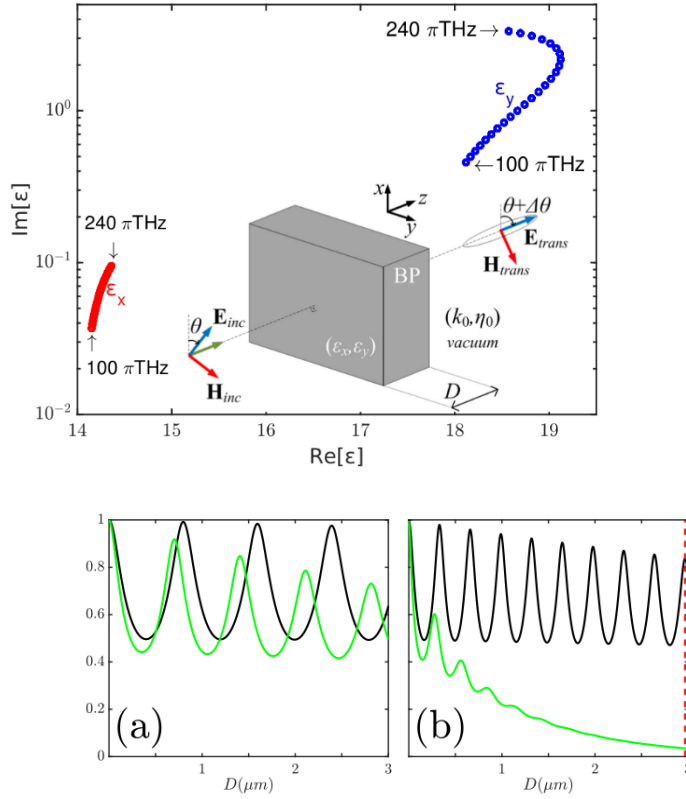


Figure 10. (a) The variation of the magnitude of the reduced transmission coefficient T_x (black) and T_y (green) as a function of thickness D for (a) $100 \pi \text{THz}$, (b) $240 \pi \text{THz}$.

Manipulating polarized light with a planar slab of Black Phosphorus:

We have studied the optical properties of Black Phosphorus (BP) monolayers stacking. First principles calculations, with Density Functional Theory (DFT), ensures that such a stacking is energetically stable. Although the dielectric function is changing dramatically as we are moving from two dimensions (single BP sheet) to three dimensions (stacking of BP sheets), after few layers the permittivity function saturates and becomes independent of the number of BP sheets. The resulting BP stacking indicates huge in-plane anisotropy in the dielectric permittivity (Fig. 10). Taking the advantage of this high anisotropy, we show that a stacking of BP can be used for polarization filtering, for tilting the polarized angle and for converting linear polarization to quasi-circular of a normal incident mixed polarized electromagnetic wave. [Manuscript submitted]

Hall effects have been guiding developments of condensed matter physics and the relation between topology and electronic structure. The conventional Hall effect involves a breaking of the time reversal symmetry, which generates a Hall response in the transport. Investigating Hall responses in other settings could be promising to elucidate and discover other aspects of the physics. A nonlinear Hall response can be generated from a system whose band structure carries non-zero Berry curvature dipole momentum near the Fermi surface. To bridge with the realistic material systems, we have performed numerical investigations of the nonlinear Hall response in various two-dimensional layered materials. The magnitude of the nonlinear response in experimental devices is in agreement with our numerical simulations.

In another approach for theoretical simulation of 2D materials, we computed the ground state electron charge density distribution inside a thin 2D layer, and then derive the electric field by solving the electrostatic Poisson equation. In the pristine crystal, the simulation and state-of-the-art TEM experiments are in excellent agreement. Similarly, the electric field can also be mapped out around defect. We are further extending our numerical simulations to these more complicated cases and to show the connection between microscopic valence bonding charge distribution and the observed electric field image.

Controlled Electrochemical Intercalation of van der Waals Heterostructures

We studied Li-ion intercalation at atomically assembled artificial van der Waals (vdW) heterostructures. We demonstrate that completely controllable new electrochemical functionality can be engineered at the vdW heterointerface that strongly enhances intercalation charge storage capacity while favorably modifying the intercalation potential for electrochemical energy storage.

Conventional approaches to preparing hybrid layered materials involve mixing component powders or solution-phase suspensions, resulting in statistical product distributions and poor control over the interface between constituent phases. In addition, the evaluation of these systems to-date relies exclusively on bulk/ensemble electrochemical measurements of these heterogeneous systems that convolute the fundamental interfacial ion insertion reactions with macroscopic transport and deleterious side reactions. Paradigm shifts in batteries, ion separation, electronics and myriad other technologies will require isolation and identification of the fundamental electrochemical characteristics of 2D heterointerfaces to enable rational materials design. To this end, we prepare individual vdW intercalation heterostructures of graphene and transition metal dichalcogenide (TMDC) layers that are encapsulated between layers of inert hexagonal boron nitride (hBN). We establish a new method, termed Hall Voltammetry, to monitor the course of the electro-intercalation reaction at the mesoscopic 2D heterointerfaces. The high quality of our 2D intercalated heterostructures permits us to use low-temperature quantum transport methods in conjunction with spectroscopy to identify the fundamental layer capacities and equilibrium potentials of individual 2D heterointerfaces for the first time (Fig. P1).

These results will open up a new research area to the physical chemistry, electrochemistry and materials science communities. By analogy to the use of functional group substituents to modify the reactivity of a molecule, or the use of specific ligands to impart unique selectivity to a mononuclear transition metal catalyst, deterministically assembling 2D layers in vdW heterostructures is a powerful way to control the coordination environment of intercalant species in novel materials. For the physics community, this study emphasizes the role of the heteroleptic vdW gap as a key degree of freedom for tuning the electric properties of a vdW material for devices with novel functionalities.

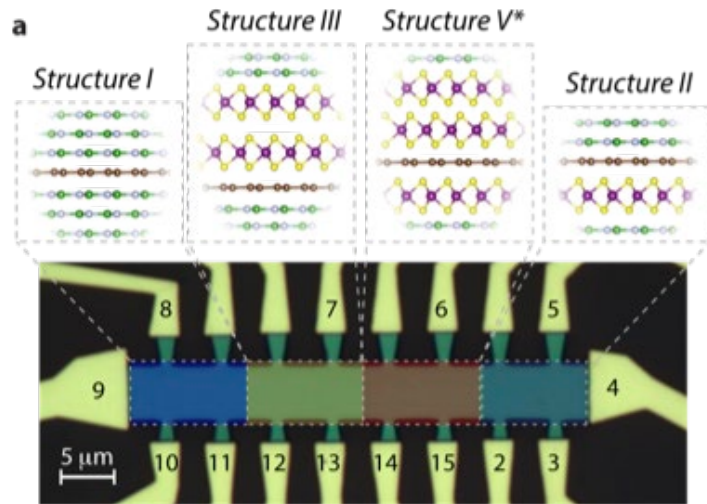


Fig. P1: Optical micrograph (false color) of a device consisting of multiple hBN-encapsulated graphene–MoS₂ heterostructure types (depicted in the associated illustration) arrayed along a single graphene monolayer.

Electrical Control of Charged Carriers and Excitons in Atomically Thin Semiconductors:

We demonstrate a complete control of charge transport in spatially confinement quantum structures fabricated in atomically thin van der Waals heterostructures. The recent development of research in 2-dimensional (2D) semiconducting materials based on semiconducting transition metal dichalcogenides (TMDCs) enables a novel engineered quantum structures. Employing functional interface realized in high-quality van der Waals (vdW) heterostructures, gate-defined electronic systems can be fabricated. In particular, spatially confined quantum structures in TMDC can offer unique valley-spin features, holding the promises for novel mesoscopic systems, such as valley-spin qubits. However, despite intense research efforts in the 2D research community, fabricating lateral quantum confined structures to demonstrate quantum transport in the TMDC nanostructures has been technically challenging due to the prevailing charge inhomogeneity in TMDCs. In this work, we successfully demonstrate the fabrication of the gate-defined quantum structures formed in atomically thin TMDC heterostructures, exhibiting quantum transport phenomena for the first time. For this work, we have developed several experimental innovations, including stamping the TMDC channel to the local gate electrodes, encapsulation of channel in 2D dielectrics, and light illumination at low temperatures, which lead to a vast improvement in the quality of the TMDC heterostructures. We report several unusual quantum transport phenomena in the TMDC heterostructures, such as the quantized conductance in gate-tunable quantum confinement in a quantum point contacts and single electron charging with tunable tunnel-coupling (Fig. P2).

We believe our work presented in this manuscript are significant for the following reasons: (i) the first experimental demonstration of completely gate controllable lateral quantum confinement structures realized in 2D materials. We demonstrate clear evidence that our method helps to improve device quality in TMDC heterostructures in both macroscopic and mesoscopic level; (ii) the experimental observation quantum transport in engineered TMDC nanostructures. Spatial confinement and manipulation of the quantum degree of freedoms of charged carriers are essential for versatile 2D quantum electronic devices.

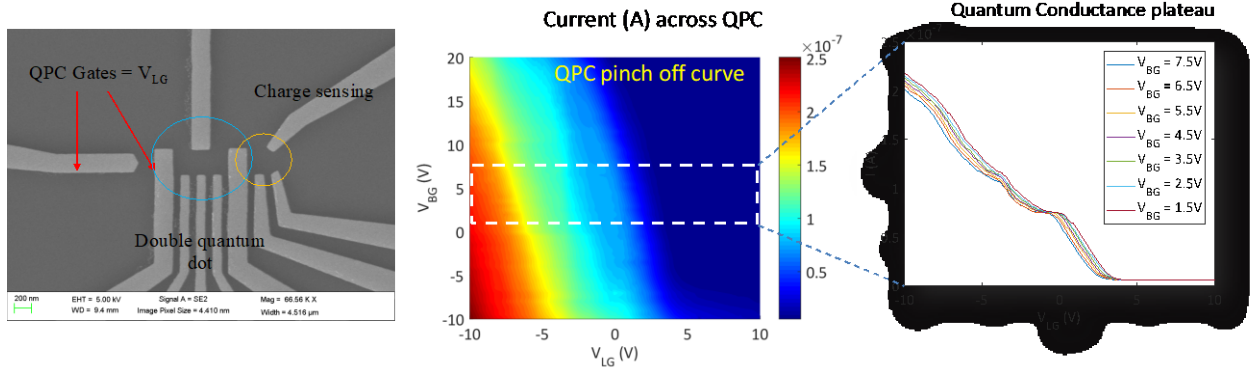


Fig. P2: By energizing the quantum dot gates (left panel), two quantum point contact channels can be pinched off over a large range of electron density with a high on-off ratio exceeding 10^6 (middle panel). The conductance changes monotonically, implying a uniform QPC channel with straight-forward gate-control. 1D cuts of the pinch-off curve at various V_{BG} reveal development of quantum plateau, a signature of quantum confinement (right panel).

Tunneling Spectroscopy of Quantum Hall States in Bilayer Graphene PN Networks:

We engineer quantum Hall edge states into forming an on-chip resonant tunneling network, and demonstrate direct measurements of their spatial profile, density of states, and compressibility in the same context with unprecedented energy and spatial resolution (see Fig. P3).

The capability of probing and engineering the spatial and energy configurations of topological states holds the key in understanding exotic electronic excitations in solid state systems and towards realizing topological quantum computing. Conventional transport studies on quantum Hall (QH) effect have been focused on demonstrating topologically protected physical observables such as quantized Hall plateaus. While it is helpful in discovering new QH excitations, these experiments are insensitive to edge state wavefunctions and local configuration of the Hamiltonian. Conventional spectroscopy tools such as scanning tunneling microscopy have been implemented, each attempted at probing a specific selection of these physical properties. In these studies, device geometry and experimental configuration prevent direct and comprehensive characterization of edge states with high spatial and energy resolutions. Utilizing electron-hole symmetry and tunable bandgap in extremely clean bilayer graphene sample, we create a unique platform allowing arbitrary placement and separation of QH edge states both in real and energy space. This allows unprecedented versatility in quantum manipulation as well as comprehensive probe of all fundamental details of QH states within the same context defined by tunable experimental conditions, providing a powerful tool and a new approach in both understanding and utilizing QH states.

The significance of our work is: (i) We demonstrate a unique electrostatically engineered system for the first time where all fundamental physical properties of the QH states are probed simultaneously in the same experimental condition, with unprecedented spatial and energy resolution; (ii) In this system, we demonstrate high degree of freedom in tailoring the spatial and energy configuration of QH states as well as equilibration between them, providing key components towards quantum manipulation, controlled braiding and advanced interferometry.

Measuring the Local Twist Angle and Layer Variation in Van der Waals Heterostructures

The properties of Van der Waals (VdW) heterostructures are determined by the twist angle and the interface between adjacent layers as well as their polytype and stacking. In this period of time, we use spectroscopic low energy electron microscopy (LEEM) and micro low energy electron diffraction (mLEED) methods to measure electronic properties of MoS₂/hBN heterostructures. We investigate results on a MoS₂/hBN heterostructure, but the methods are applicable to other materials. Diffraction spot analysis is used to assess the benefits of using hBN as a substrate. In addition, by making use of the broken rotational symmetry of the lattice, the cleaving history of the MoS₂ flake is determined, that is, which layer stems from where in the bulk.

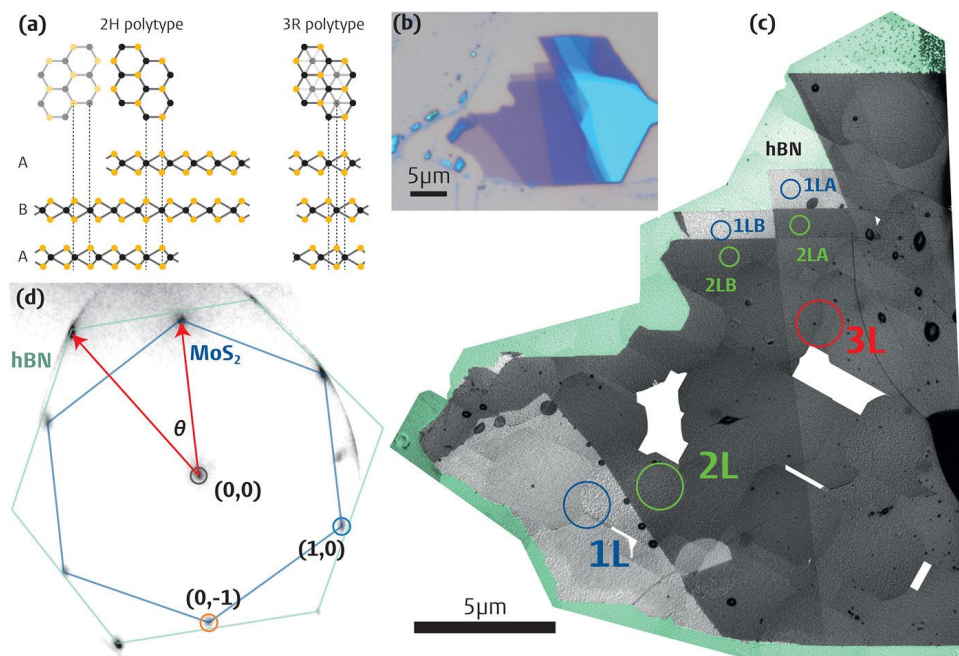


Fig. P1: (a) Top view and side view of the atomic structure of the 2H and 3R polytypes of MoS₂. b) Optical microscope image of the MoS₂ flake on SiO₂ before transfer, showing a clear layer contrast between different layer counts. c) Overview of the MoS₂ flake composed of 90 bright-field LEEM micrographs. The MoS₂ flake shows different intensities, the surrounding hBN substrate is shaded green for clarity. d) mLEED measurements with both hBN as well as MoS₂ in the aperture. From this we determine the twist angle θ between the hBN and the MoS₂.

Imaging quantum dot formation in MoS₂ nanostructures

Among two-dimensional materials, semiconducting ultrathin sheets of MoS₂ are promising for nanoelectronics. We show how a scanning probe microscope (SPM) can be used to image the flow of electrons in a MoS₂ Hall bar sample at 4.2 K allowing us to understand device physics at the nanoscale. The SPM tip acts as a movable gate and capacitively couples the SPM tip to the device below. By

measuring the change in device conductance as the tip is raster scanned across the sample, spatial maps of the device conductance can be obtained. We present images showing the characteristic ‘bullseye’ pattern of Coulomb blockade conductance rings around a quantum dot formed in a narrow contact as the carrier density is depleted with a backgate. These images show that multiple dots are created by the disorder potential in MoS₂. From these SPM images, we estimate the size and position of these quantum dots using a capacitive model.

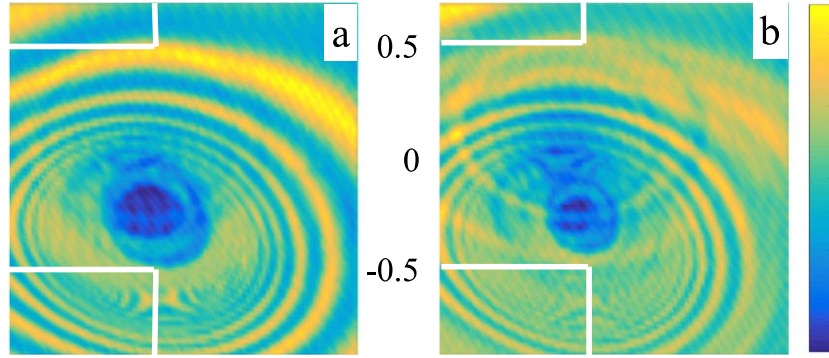


Fig. P2: (a) Display of conductance change ΔG versus tip position at the narrow contact (see figure 1) for $V_G = 4.80$ V, when the device is nearly depleted. The bullseye pattern of concentric rings are Coulomb blockade conductance peaks associated with a quantum dot at the center. (b) As the density is increased for $V_G = 5.29$ V an additional quantum dot appears at a different location.

Superconductivity in twisted trilayer graphene

We discovered enhanced superconductivity that can be tuned by electric field effect in twisted trilayer graphene, providing compelling evidence for unconventional superconductivity in this system. Engineering moiré superlattices by twisting and stacking two layers of Van der Waals materials has proved to be an effective way to promote interaction effects and induce exotic phases of matter. After the discovery of superconductivity and correlated insulators in magic-angle twisted bilayer graphene (MA-TBG), several different two-dimensional materials have been utilized to create two-layer twisted systems and various novel phases. However, superconductivity, which sparked much of the initial excitement, is still not well understood, nor has it been conclusively realized in a twisted system other than MA-TBG. Extending the recent theoretical prediction, here, we present an experimental realization of magic-angle twisted trilayer graphene (MA-TTG), in which we observe both electron density and electric field tunable superconductivity. Utilizing the electric-field controlled electron energy band in MA-TTG, we demonstrate enhanced superconductivity. We also discover that the origin of superconductivity stems from strong interaction, which cannot be described by conventional BCS type of superconductivity.

The significance of our work is to demonstrate that the electric field tunability allows for investigation of the relationship between flat band structure and superconductivity by tuning the band structure in situ

with the electric field, a crucial step to understanding the origin of moire superconductivity. Moreover, should the superconductivity be interaction driven, twisted trilayer graphene intrinsically has stronger Coulomb interaction due to larger magic angles, and therefore may provide a way to enhance the superconductivity.

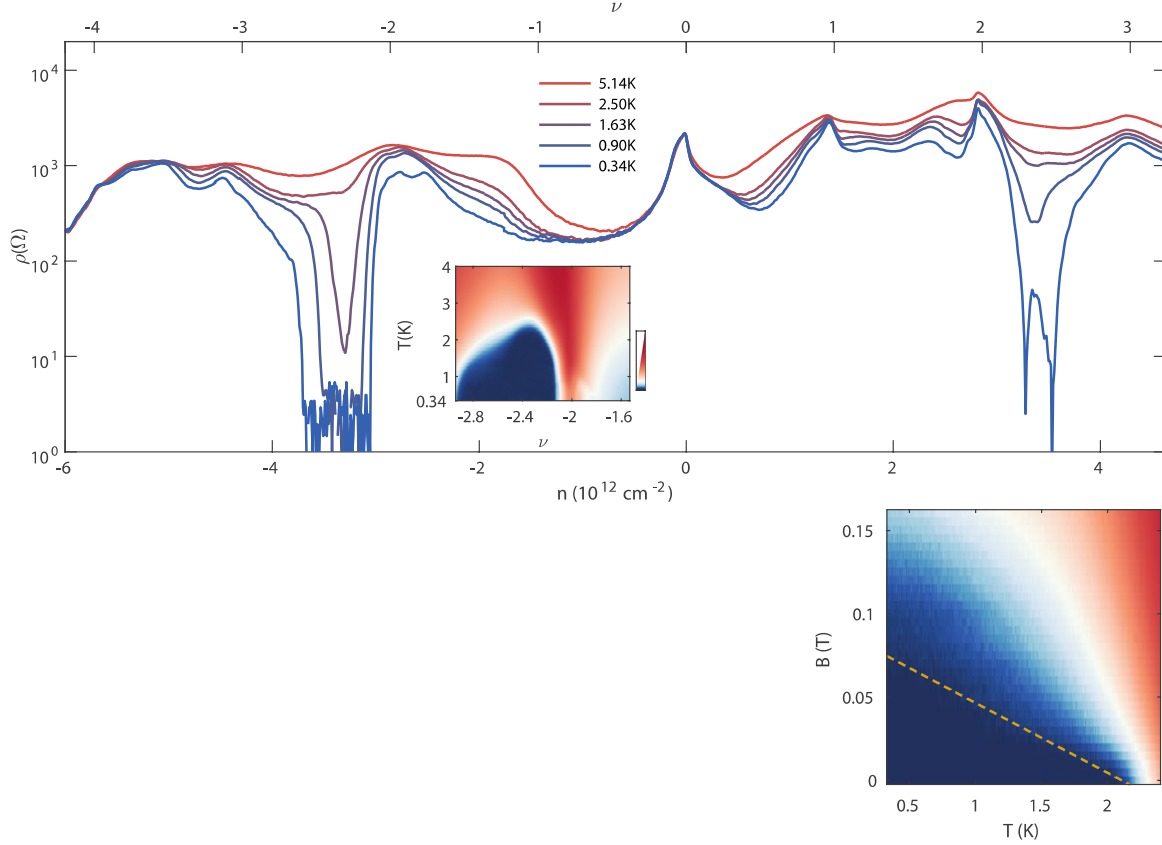


Fig. P1: Superconductivity in TTG. (a) resistivity as a function of moire band filling at several different temperature values. The left (right) inset shows the superconducting domes. (b) Superconducting transition in resistivity. The BKT transition temperature is marked where $\nu \sim T^3$ as shown in the inset. (c) Differential resistance measurements as a function of bias current at different magnetic fields. (d) resistivity as a function of temperature and magnetic field. The dashed line corresponds to a GL theory fit.

Correlated Insulating States and Superconductivity in twisted trilayer graphene Moire of Moire Superlattice

In the first stage of MURI add-on, we have reported [16] correlated insulating states near the half-filling (fig. 1a) of the moiré of moiré superlattice at an extremely low carrier density ($\sim 10^{10} \text{ cm}^{-2}$), near which we also report zero-resistance transport behaviors typically expected from a 2D superconductor (fig.1b,c,d), with critical current and critical temperature characterized to be several times larger than previously reported tBLG systems. Moreover, the temperature dependence of the measured resistances

at full-occupancy states are semi-metallic, distinct from the insulating behavior of the twisted bilayer systems, providing a first demonstration of the emergent correlated transport behavior from continuous, non-isolated moiré flatbands. Our findings shed new insights into unraveling the microscopic mechanisms of moiré correlated states and provide the basis for future studies on this exciting new material platform, such as demonstration of superconducting phase coherence and utilizing it for quantum electronics devices and computing platform.

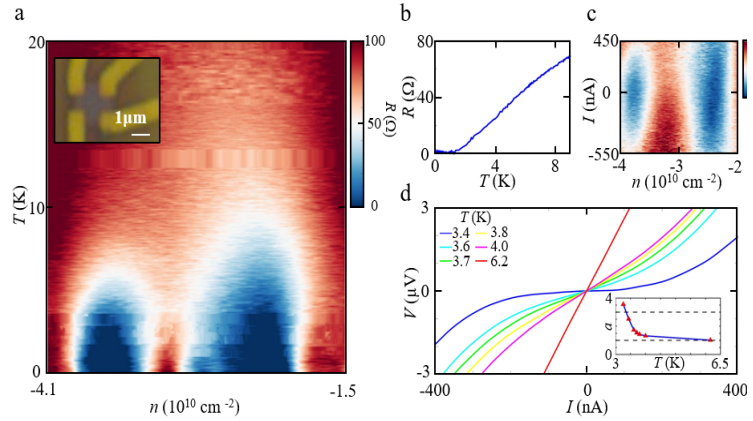


Fig K1. tTLG superconductivity demonstrated during this MURI add-on. (a) 4-probe resistance measurement of tTLG sample (inset) at variable low temperature, showing co-existing correlated insulator and superconducting states at extremely low carrier density on the order of $\sim 10^{10} \text{ cm}^{-2}$. (b) Typical transport behavior expected from 2D superconductor with (c) critical current and (d) critical temperature characterized to be several times larger than previously-reported tBLG.

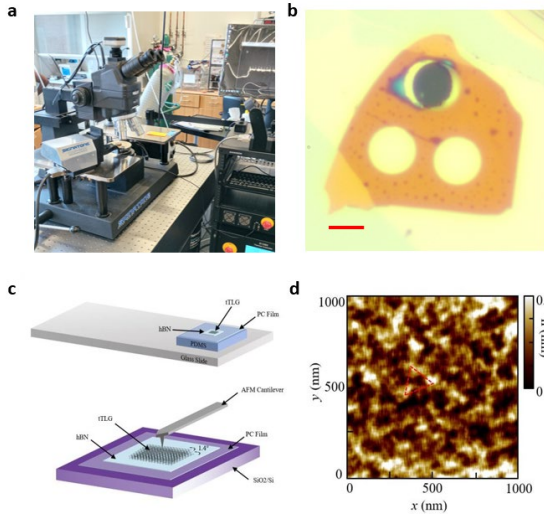


Fig K2. Improved Experimental Capability in Fabricating Advanced tTLG Heterostructures. (a) Newly configured dry transfer stage with more precise control over twist angle and atomic strain, allows assembly of specialized tTLG stack including (b) suspended tTLG stack with ultra-thin hBN capping layers for TEM study and (c)(d) mechanically-flipped partially-encapsulated tTLG stack for directly characterization of Moire of Moire lattice topography via AFM.

Structural Study of twisted trilayer graphene Moire of Moire Superlattice

Over the past year's MURI support period, the experimental focus has been on the direct structural study of tTLG, particularly the twist-angle inhomogeneity and mechanical relaxation responsible for a plethora of Moire of Moire length scale, critical in understanding the rich underlying physics of tTLG and reliably harness its unique quantum phenomena for next generation quantum devices. To this goal, Wang lab have newly acquired and fully configured an advanced dry-transfer stage (Signatone) with improved the control over twist angle (fig. K2a). This allowed us to develop a series of recipe over the past year for assembly of more advanced specialized tTLG stacks designed specifically for spectroscopy measurement. For example, we have developed a method to consistently flip the tTLG stack without introducing additional atomic strain (fig. K2c), allowing the Atomic Force Microscope (AFM) tip to get in close proximity to the tTLG surface and directly characterize [16] the topography of Moire of Moire lattice (fig. K2d). In addition, in

collaboration with Kim group (Harvard), we have developed and fabricated multiple tTLG stacks with ultra-thin hBN capping layer, suspended over pre-etched openings on a SiN substrates. These samples have recently been shipped to Kim lab for advanced Tunneling Electron Microscopy (TEM) study (fig. K2b).

Development of Gate-defined Quantum Confinement for Graphene

We have also made significant progress in electrical transport measurement of tTLG. First and foremost, we have developed a dual-bottom-gated quantum point contact that allows sensitive control over tunnel coupling between adjacent 2D regions of any gapless vdW materials (fig. K3). This provides an important technical component towards the gate-defined Josephson junctions based on Moire superconductivity in tBLG and tTLG. Moreover, we have fabricated and measured dual-graphite-gated tTLG samples, which preliminarily demonstrated gate-tunable electron correlation as a function of applied displacement field, shedding new lights on the rich underlying correlated physics and providing new experimental tuning knobs for tTLG quantum devices.

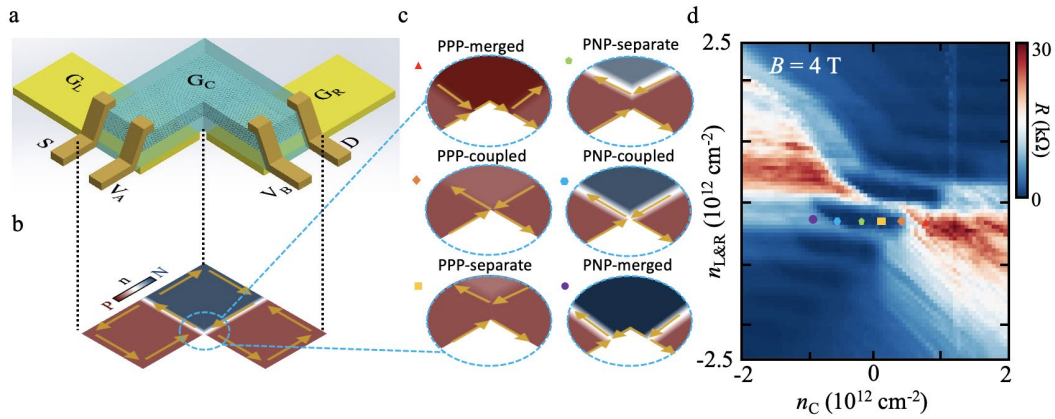


Figure K3 | Dual-bottom-gated Quantum Point Contact for Tunable Coupling in Gapless 2D Material. (a) Device Schematic of monolayer graphene device. The device is etched into a “Λ” shape to form a QPC at the junction point of the three regions controlled by G_C, G_L and G_R. (b) Color plot maps the carrier densities in all three regions. (c) The coupling between difference regions of the device can be sensitively tuned to different transport regimes. (d) Phase diagram of the QPC under a magnetic field of 4T as a function of n_C, n_L and n_R. Zero and finite resistance regions correspond to merge and decouple regimes respectively.

Probing Interlayer Screening and Moire Band Evolution via Gate-defined PN junctions

When a conventional twisted-bilayer Hall-bar is measured as a whole, exotic quantum electronic behaviors can emerge when specific material conditions are met, often in selected few devices with relatively low experimental yield. Large sample-to-sample variation has been reported, due to the extreme sensitivity of the system to layer-specific details, such as lattice relaxation, atomic strain, interlayer screening, and resulting charge imbalance and band reconstruction. Experimental approaches that comprehensively characterize these materials parameters can therefore be a key component of understanding the microscopic mechanisms of moiré physics and developing more robust and versatile control over future twistrionic devices.

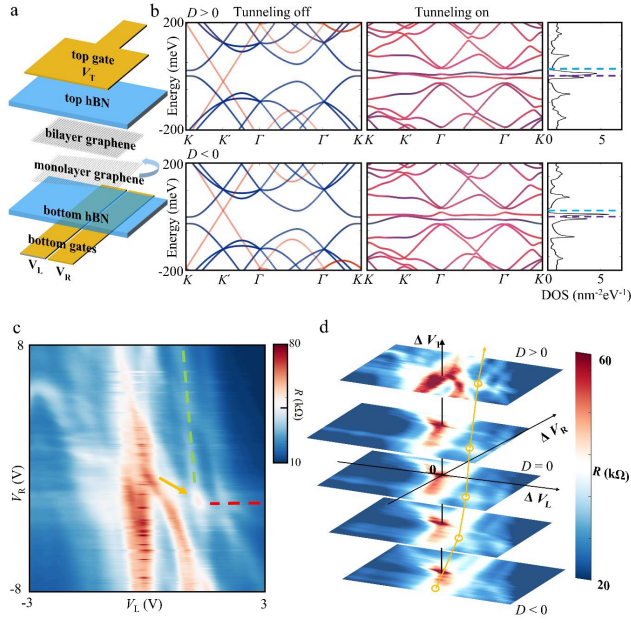


Figure K4. Probing Moiré Band Evolution due to Interlayer Screening with tMBG PN junction. (a) Schematic diagram of a tMBG device. (b) Calculated bandstructure of tMBG system at positive (top panels) and negative (bottom panels) displacement field before (left panels) and after (middle panel) turning on the interlayer coupling and corresponding DOS (right panel). (c) (d) Measured 4-probe resistance as a function of the adjacent gate voltages V_L and V_R , showing a series of high resistance peaks, which correspond to left (green), right (red) and both (orange) doped (resonance) at local DOS minima, characterizing evolution of moiré band due to interlayer-screening.

over the qualitative behavior of the PN junctions by controlling the layer polarization. By using the same device architecture in a twisted monolayer-bilayer graphene (tMBG), we control the energy at which moiré band-crossing occurs, by tuning the bilayer graphene band structure while using the monolayer graphene as the reference (fig.K4). High resistance peaks were observed when both P and N side of the PN junction are doped near the avoided crossing, which we use to characterize band evolution as a function of displacement field.

However, layer-specific electrical properties remain largely unexplored, because conventional magneto transport measurements of Hall bar devices reveal electronic properties of the system as a whole instead of each individual layer. To characterize these parameters and provide proof-of-principle demonstration of controlling them, we have fabricated gate-defined PN junctions on two different twisted bilayer systems: tDBG and tMBG. The material combination has specifically been chosen for experimental access to layer specific details: tDBG allows characterization and control of the layer polarization and inter-layer screening, while tMBG provides an ideal material combination for tuning the individual band evolution and the resulting moiré-band crossing. We developed [17] a device architecture for twisted bilayer systems consisting of two atomically flat bottom gates and a single global top gate that allows precise characterization and control over layer-specific material parameters. By measuring electrical transport across the gate-defined PN junctions in twisted-double-bilayer graphene (tDBG), we characterize the layer polarization and electrostatic screening in each composing layer and demonstrate versatile control

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